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Quantification of Indoor Air Pollution from Using Cookstoves and Estimation of Its Health Effects on Adult Women in Northwest Bangladesh

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ABSTRACT

A longitudinal stove intervention study was conducted in Northwest Bangladesh between August 2005 and June 2009 to quantify the reduction of indoor air pollution from a Bangladeshi manufactured and commercialized cookstove (the BCSIR improved stove). In the post-intervention phase of the study, the indoor particulate matter (PM) size distribution and chemical composition, as well as carbon monoxide (CO) concentration, were characterized in 40 kitchens, 50% with the BCSIR improved stoves and 50% with traditional stoves. The TSI DustTrak, P-TRAK and Q-TRAK Plus, along with the UCB Particle Monitor and the Onset HOBO, were simultaneously deployed to continuously characterize Carbon Monoxide (CO) and Particulate Matter (PM). Detailed chemical composition was quantified from X-Ray Flouroscence and Carbon Analyzer. Median 24-hr concentrations for CO and $PM_{2.5}$ were 2.5 mg/m^3 and 1.8 mg/m^3, respectively, for the unimproved mud stove kitchens and were 2.0 mg/m^3 and 0.73 mg/m^3, respectively, for the BCSIR improved stove kitchens. These differences were equivalent to 23% and 59% reduction of CO and $PM_{2.5}$ concentrations, respectively. The cook's daily exposure was estimated from these measurements to assess health impacts. Ultrafine particle number concentrations were 15,000 ± 7,200 pt/cm^3 during non-cooking periods and 75,000 ± 31,000 pt/cm^3 during cooking periods. Of the chemical composition of the $PM_{2.5}$ emitted from cooking, 59–60% was organic matter and 29–30% was elemental carbon. The predominant chemical species were elemental carbon (EC), organic carbon (OC), chlorine, and potassium. These results demonstrate possible reduction of PM and CO from cooking with improved stoves in rural areas in Bangladesh where solid fuels are used for cooking.

***Keywords*:** $PM_{2.5}$; Carbon monoxide; Wood burning; Combustion aerosols; Personal exposure; Indoor air pollution.

INTRODUCTION

One of the World Health Organization (WHO)'s millennium goals is to improve health of the rural poor, emphasizing improved cook stove installations to reduce exposure to harmful air pollutants (WHO, 2009). Indoor air pollution (IAP) from poor quality household fuels in open fires or poorly designed stoves in developing countries has been linked to acute respiratory infection (ARI), chronic obstructive pulmonary disease (COPD), and lung cancer (Smith *et al.*, 2000). Acute respiratory infections, primarily pneumonia, are the leading causes of morbidity and mortality in very young children in Bangladesh accounting for approximately 25% of all deaths in children under five and 40% of deaths in infants (Baqui *et al.*, 1998). A large fraction of the 150 million people living in Bangladesh employs traditional three stone fires, using various mixtures of solid fuels such as biomass, coal, animal dung, crop residues, typically rice straw and dried leaves. To quantify air pollution from burning the above mentioned solid fuels in unimproved mud stoves, Dasgupta *et al.* (2006 and 2009) measured particulate matter with aerodynamic diameter less than or equal to 10 μm in diameter (PM_{10}) in the Narayanganj District of Bangladesh. In order to improve fuel efficiency and reduce indoor air pollution levels, the Deutsche Gesellschaft für Technische Zusammenarbeit (GTZ), the German development organization, and Grameen Shakti have been leading the efforts in improved stove

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installation in various locations in Bangladesh. Despite these efforts, no detailed measurements have been published in Bangladesh to quantifying the indoor air pollution levels in households before and after stove installation, thus making the quantification of health improvement from IAP reductions by these stove intervention projects very difficult.

In the Saidpur-Parbatipur municipalities of northwest Bangladesh a longitudinal stove intervention study was launched by Winrock International and United States Agency for International Development (USAID) as part of a household energy project. The objective of this project was to develop and demonstrate how an integrated approach in stove technology dissemination could lead to behavioral changes in consumers, leading to improved energy efficiency and reduced indoor air pollution in the households. The technology of choice was based on an improved stove designed by Bangladesh Council of Scientific and Industrial Research (BCSIR) with newly trained technicians within the project location. These improved stoves were promoted and disseminated in the region. Viable partnerships were developed with local artisans and business owners for sustained demand and repair of the improved stoves. Within this larger project, the effects from the introduction of the stoves in targeted homes to indoor air pollution level change was monitored and quantified. The results of this IAP study were collected during June 2009 and are presented here. In this study, we utilized several state-of-the-art instruments to monitor IAP using a cross-sectional study approach. We also explored the CO to PM relationship and compared our findings with other similar studies conducted in other locations around the world.

METHODS

Study Site Description

During June 2009, a cross section of forty households was selected. Half of the study households owned and regularly cooked with a BCSIR improved stove (intervention group) and the other half cooked with a traditional mud stove (control group). The BCSIR stoves are improved stoves with an iron grate and a chimney to conduct the flue gases outside the house through a hole in the roof. Fig. 1 shows the improved and traditional stoves with design parameters as well as views of the primary and secondary fuels used in this study. Detailed description of this stove can be obtained in the supplemental method section. When selecting households from the study site, a homogeneous community was considered to ensure minimum differences in household socio-economic status, neighborhood characteristics, and local culture and customs. For this reason, the cross-sectional study was conducted in a single community located within two adjacent wards (Ward # 13 and 14) in the Saidpur location in Northwest Bangladesh. The map of the study site can be found in Fig. 2. In this particular location, most of the households were built close together or attached with each other, separated by a wall.

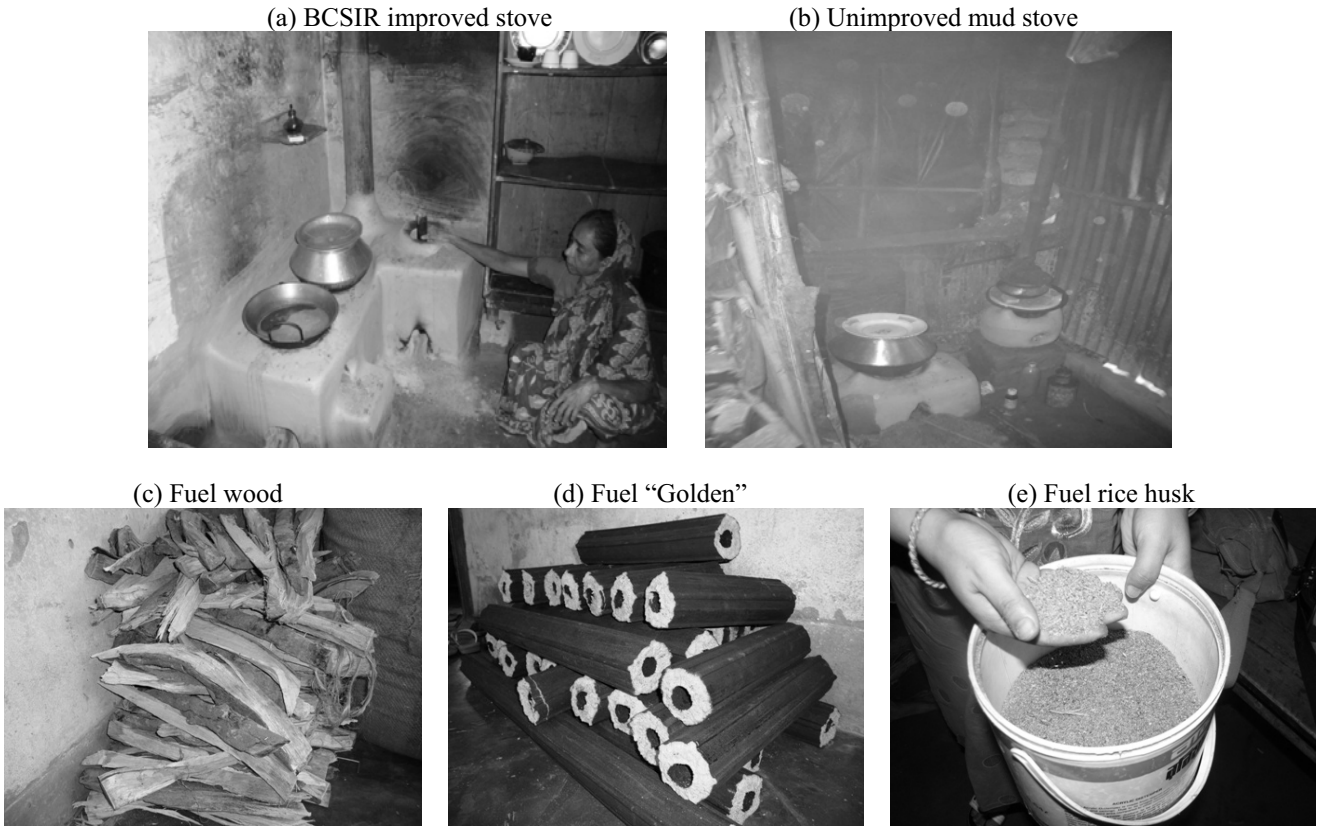


Fig. 1. Photos of (a) the BCSIR improved stove, (b) unimproved mud stove, (c) primary fuel wood, (d) secondary fuel "Golden", and (e) fuel for lightening the fire, rice husks.

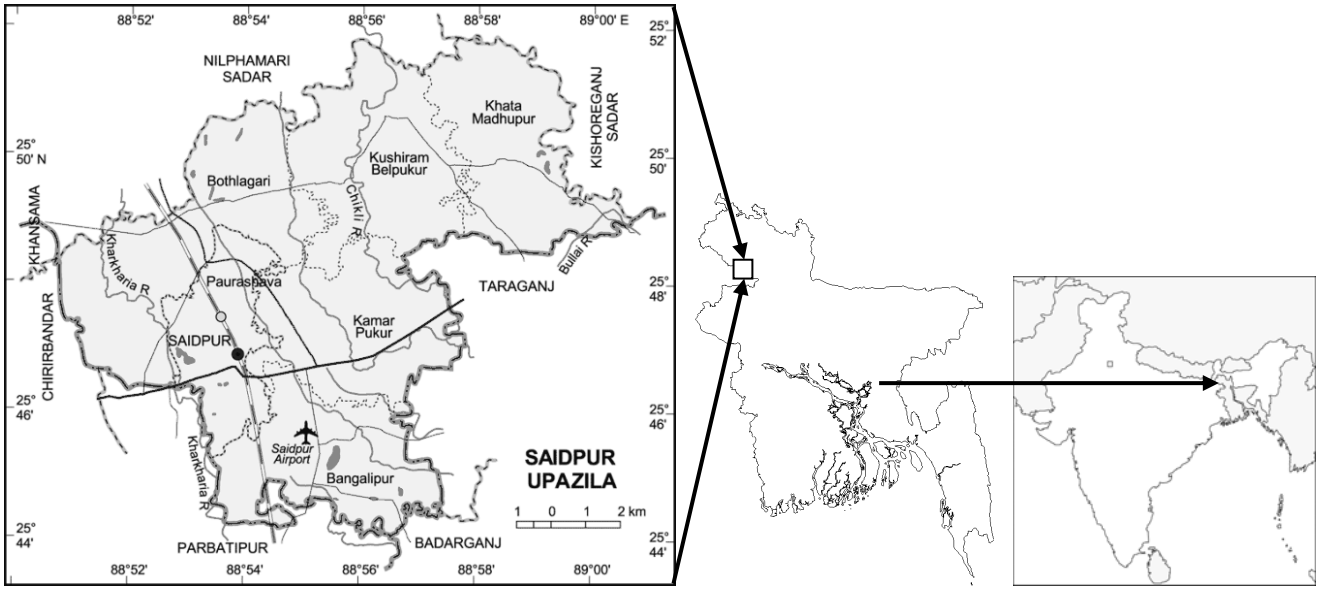


Fig. 2. Map of the study area in Saidpur Upazila in Bangladesh.

Tables 1 and 2 compare characteristics between the BCSIR improved stove (intervention) and traditional mud stove (control) households. The wall and roof of the households were mostly constructed with brick and tin, respectively, while a minority of the households used a mixture of bamboo and brick as the construction material. Primary and secondary fuel sources were similar in nature, consisting of wood and woodchips, "Golden", and other (cow dung and/or leaves). "Golden" is a popular source of fuel made of compressed rice husk and shaped into a hollow log with binding glue. Before use, the hollow log pieces were sliced vertically into circular pieces and fed into the cook stove as fuel. As a primary fuel, wood and "Golden" were predominantly used and purchased from local markets. Most households used only a single source of fuel (see Table 1, primary fuel). For those households using supplemental fuel (secondary fuel), the choices were among wood and "Golden" with some houses using other (cow dung and/or leaves). Potential combustion sources other than the cook stove were also quantified with the aid of surveys. As seen in Table 1, electricity was the main lighting source in the kitchen with 3 houses in both the control and intervention groups using kerosene to light their kitchens.

Some household characteristics were quantified and statistically compared according to stove status as shown in Table 2. Some of these household physical traits represent the ventilation of the houses as well as quantification of the fuel use and cooking duration. A t-statistic was calculated for each of these variables, and as seen from Table 2, the control homes were not significantly different from the intervention homes. This similarity reduces the number of variables that can potentially lead to differences in IAP levels.

Table 1. Characteristics of participating households.

Variables	Description	Control (N = 19)	Intervention (N = 21)
Wall Construction	Bamboo	2	6
	Brick	11	13
	Tin	0	1
	Bamboo & Brick	6	1
Roof Construction	Bamboo	4	3
	Brick	1	4
	Tin	14	14
Kitchen Lighting	Electric	17	19
	Kerosene	2	1
	None	0	1
Primary Fuel	Wood	11	10
	Golden	7	9
	Others	1	2
Secondary Fuel	Wood	5	6
	Golden	3	5
	Others	1	4
	None	10	6

Table 2. Statistical comparison of the characteristics of participating households.

Stove Status	Statistical Parameters	# Wall	Holes in Wall	# Door	# Window	Fuel Use/Day in Kg	Secondary Fuel Use Per Day in Kg	# People Cooked for	Hrs Cooked	Hrs Secondary Stove
Control	Minimum	2	0	1	0	0	0	2	2.5	0
	Maximum	4	5	3	4	15	0.5	8	7	0
	Mean	3.9	2.0	1.8	0.6	3.6	0.3	5.9	4.0	0
	Standard Deviation	0.4	1.5	0.8	0.9	3.5	0.3	1.8	1.4	0.0
	Range	2	5	2	4	15	0.5	6	4.5	0
	N =	19	19	19	19	17	19	19	18	19
Intervention	Minimum	2	0	0	0	0	0	2	0.5	0
	Maximum	4	5	4	2	10	2	8	9	1
	Mean	3.8	1.8	1.5	0.6	3.2	0.5	5.9	4.2	0.1
	Standard Deviation	0.5	1.4	1.0	0.7	2.1	0.5	2.1	1.9	0.2
	Range	2	5	4	2	10	2	6	8.5	1
	N =	21	21	21	21	20	20	21	21	21
P-Value (CI = 95%)		0.377	0.598	0.275	0.912	0.654	0.071	0.991	0.701	0.214

Monitoring of CO and $PM_{2.5}$

The IAP monitoring was divided into two components: intensive and extensive monitoring. In the intensive monitoring, a wide-range of instrumentation was co-located with duplicate sets for four consecutive days in two households, one house with the BCSIR improved stove and the other house with the traditional mud stove. In the extensive monitoring, two instruments were used for a single day in each house covering a total of 40 households, 50% of the houses with the BCSIR improved stoves and other 50% of the houses with traditional mud stoves. The purpose of the intensive monitoring was two-fold: 1) instruments were co-located to check the reliability and accuracy of the instruments for cross-calibration, and 2) detailed chemical characterization of particulate matter and carbon monoxide was targeted to better understand the emission characteristics from the stoves. However, the purpose of the extensive monitoring was to statistically quantify fine particulate matter and carbon monoxide concentrations in the kitchens from using the stoves. For both type of monitoring, all household owners/cooks were requested to use their primary stove as much as feasible without compromising their cooking pattern, fuel use, or their usual behavior.

The intensive monitoring sampling scheme used 22- to 25-hr long measurements, repeated over four days for each stove type. Each sampling day captured the cooking events (lunch, dinner, and following morning's breakfast). The improved stove house was monitored from June 21 to June 24, 2010 and the traditional stove house was monitored from June 25 to June 28, 2010. Two TSI DustTraks (Model 8520), two TSI P-TRAKs (Model 8525), two TSI Q-TRAKs Plus (Model 7565), two $PM_{2.5}$ gravimetric instruments (SKC Aircheck XR5000 with BGI SCC1.062 Triplex Cyclone), two University of California at Berkeley Particle Monitor (known as, UCB, Model P3; Litton *et al.*, 2004; Chowdhury *et al.*, 2007a), and two Onset HOBOs were co-located in each house each day, providing continuous measurements for $PM_{2.5}$ and CO, as well as integrated $PM_{2.5}$ obtained gravimetrically. The inlets of all the devices were hung approximately 3 inches apart from each other and the instruments were placed on a shelf fixed to the wall. The height of the shelf was 1.4 meter and 1.0 meter from the floor for the improved stove kitchen and traditional stove kitchen, respectively. The distance from the center of the stove to the instruments was kept between 1 and 1.5 meter. In the traditional stove kitchen, a porous bamboo wall separated the devices from the stove. This arrangement ensured the security of the instruments during the 4 days of sampling since the kitchen had a 24-hr open entryway allowing free movement of people from outside. Details of the instrument protocol and measurements are provided in the supplemental method section. In addition to these indoor measurements, a single gravimetric instrument measuring $PM_{2.5}$ was deployed outside the house to measure the neighborhood air pollution.

The extensive monitoring sampling scheme with 22- to 25-hr single measurement for each of the 40 households captured all cooking events within each sampling period.

$PM_{2.5}$ and CO were measured continuously and recorded every minute during each sampling period using the UCB and the Onset HOBO, respectively. These instruments were used in the extensive monitoring because of their noiseless operation and compact size. A questionnaire was administered during both the intensive and extensive monitoring periods to record details about the conditions in the house and the sampling instruments. This questionnaire was conducted for every household immediately at the end of sampling. Questions about cooking events, household use of lamps, smoking, trash burning, and IAP monitor removal or displacement were asked to the cook(s). The results obtained from these surveys are presented in Tables 1 and 2.

QA/QC

Two field workers were trained to conduct the sampling. To ensure quality of the data, a supervisor visited approximately 12 hours after the start of the sampling. This approach allowed the field workers to check the instruments to ensure that they were not compromised. Eight duplicates (67% of the total samples), nine field blanks, and ten lab blanks were utilized to assess the repeatability and detection limits of the PM concentration measurement method, as well as for possible filter contamination. The duplicate and field blank filters were included in the measurements throughout the whole fieldwork. Eight duplicate(17% of the total samples) UCB to UCB collocated with two DustTraks were used to measure $PM_{2.5}$ concentration and eight (17% of the total samples) HOBO to HOBO collocated with two Q-TRAKs Plus were used to measure CO concentration in the intensive homes. All field blanks were handled in the same manner as the sample filters but no air was pulled through the filters. The average gained weight for all blanks was 0.002 mg which was used to blank correct the samples. During the entire study period, the same rotameter was used in the field. The flow rates from the rotameter were 1.5 ± 0.02 L/min and 1.5 ± 0.03 L/min for the initial and the final measurements. Because of this flowrate accuracy, the size cut was unaffected from their target values.

Chemical Analysis

The Teflon and the quartz filters, deployed indoor and outdoor during the intensive monitoring, were analyzed for chemical species present in $PM_{2.5}$. Elemental carbon (EC), total carbon (TC), and organic carbon (OC) were analyzed from the quartz filters by the Sunset Laboratories OC/EC instrument (Sunset Laboratories, OR, USA) at the Air Quality Laboratory at SDSU. The Teflon filters were shipped to Clarkson University where X-Ray Fluorescence (XRF) was conducted to quantify up to 42 trace metals.

Data Analysis and Validation

As described in Chakrabarti *et al.* (2004), relative humidity (RH) corrections were applied to each minute of UCB and DustTrak data to account for increased light-scattering caused by high RH. RH during the study ranged between 41%–89% with no significant difference of RH between the type of stoves. We employed the RH correction equation from Chakrabarti *et al.* (2004) as follows, $1 + ((0.25RH^2)/(1 - RH))$. Minute-by-minute RH was obtained from the Q-TRAK Plus and HOBO measurements. Additionally, all instrumentation was calibrated according to field-based calibration method presented in Chowdhury *et al.* (2007a). These field-based calibrations were conducted using the intensive monitoring dataset where co-located measurements were present from different instruments, measuring the same pollutant. For example, fine particle ($PM_{2.5}$) gravimetric measurement were used as "gold standard" to correct the data from the light scattering devices, such as UCB and DustTrak, to account for changes in light scattering due to size distribution and particle color (McNamara *et al.*, 2011). Also, the TSI Q-TRAK was used to correct the data from the co-located HOBO. Finally, the slight differences between the UCB and DustTrak in processing the second-by-second samples that were averaged and logged into minute-by-minute data caused one to two minutes of delay between the instruments. To correct this slight discrepancy, the DustTrak dataset were shifted by 1 to 3 minutes to synchronize them with the UCB start time. Once these three-step corrections were completed, scatter plots were constructed with the cleaned dataset to show the correlation between related instruments, measuring the same pollutant. Five outlier data points were identified and removed based on evidence from the field notes and household questionnaire responses. Any unusual cooking activity or instrumental failures were identified and data removed accordingly. The high correlation achieved using the cleaned data suggests the validity of the measurements during intensive monitoring.

RESULTS

Time-series graphs of the minute-by-minute $PM_{2.5}$ concentrations for the BCSIR improved stove and the unimproved mud stove kitchens measured on two selected days (6/21/2009 and 6/28/2009) are shown in Figs. 3(a) and (b), respectively. These graphs show the periods when $PM_{2.5}$ concentrations peak, corresponding to different cooking times. The breakfast and lunch preparation times can be seen in Fig. 3(a) and how these two meals were combined into one cooking period in Figs. 3(b). Fig. 3(c) show the concentration of CO over 24 hours measured by co-located HOBO and Q-TRAK Plus, along with $PM_{2.5}$ concentrations. There is also a strong correlation between the UCB and DustTrak used to measure $PM_{2.5}$ in the kitchens with the improved and un-improved stoves on 06/21/2009 and 06/28/2009, respectively (see Figs. 3(a) and (b)). The scatter plots in Figs. 4(a) and (c) reflects the correlation between the UCB and DustTrak as seen in Figs. 3(a) and (b), respectively. If the high points were removed from Fig. 4(a), the new linear equation changes to $y = 1.2x + 0.038$ with an $r^2 = 0.64$ which is higher than the current $r^2 = 0.62$.

Fig. 4(b) show the correlation ($R^2 = 0.79$) between data values of CO concentration, measured by co-located HOBO and Q-TRAK in households (HH) during the intensive monitoring. CO concentrations in ppm have been converted into mg/m^3 following the method outlined in Northcross *et al.* (2010). Table 3 presents the descriptive statistics for $PM_{2.5}$ and CO concentration by stove status. These values,

calculated from the extensive study, provide a measure of the overall exposure of the HHs in this sample size.

Ultrafine Particulate Matter

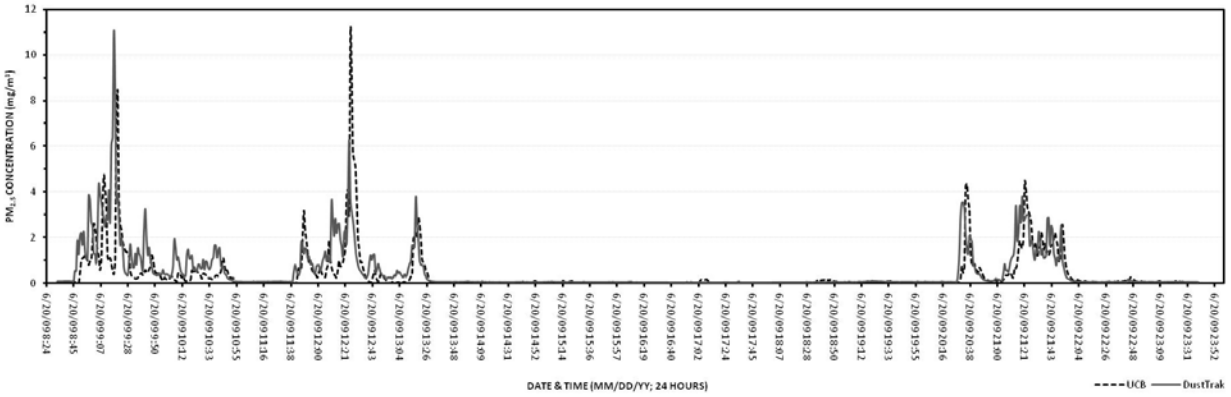
In the present study, number concentrations of ultrafine particulate matter (0.02 to 1.0 micron diameter) were measured by using two TSI P-TRAKS. To date, no ultrafine data is available in any locations in Bangladesh. During cooking and non-cooking periods these concentrations were 75,000 ± 31,000 pt/cm^3 and 15,000 ± 7,200 pt/cm^3, respectively. These levels are much higher than typical concentrations seen near busy highways in the US. This finding suggest that ultrafine PM emissions from biomass cookstoves can potentially penetrate very deep into the respiratory and cardiovascular system.

DISCUSSION

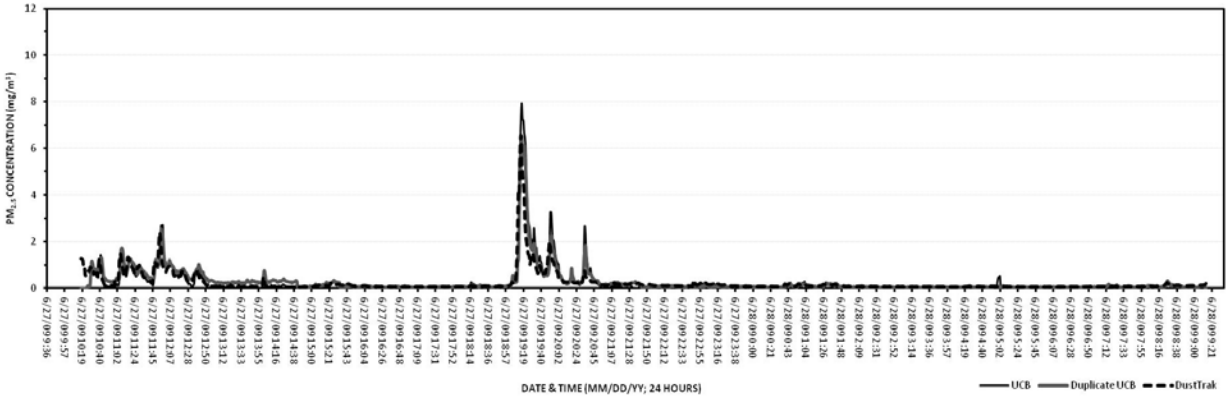
CO to $PM_{2.5}$ Correlation

Correlations between $PM_{2.5}$ and CO in the extensive monitoring, seen in Fig. 5, show r^2 of 0.61 (N = 15) for unimproved mud stove kitchens and 0.71 (N = 16) for the

Panel 3(a): June 21, 2009 ($PM_{2.5}$)



Panel 3(b): June 28, 2009 ($PM_{2.5}$)



Panel 3(c) : June 21, 2009 (CO and $PM_{2.5}$)

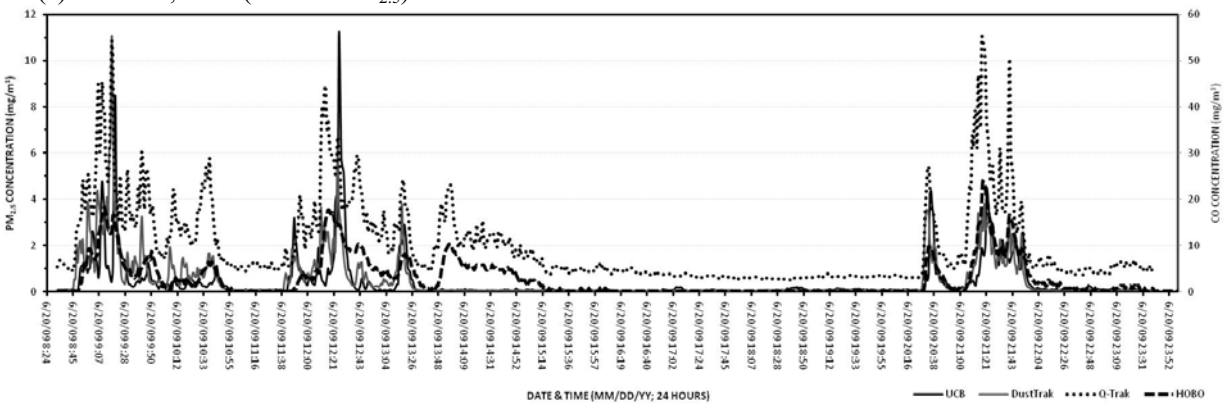


Fig. 3. $PM_{2.5}$ concentrations (mg/m^3) measured by UCB, duplicate UCB, and DustTrak, and CO concentrations (mg/m^3) measured by HOBO and Q-TRAK Plus in kitchens with the BCSIR improved stove measured on June 21, 2009 [panels (a) and (c)] and unimproved mud stove on June 28, 2009 [panel (b)].

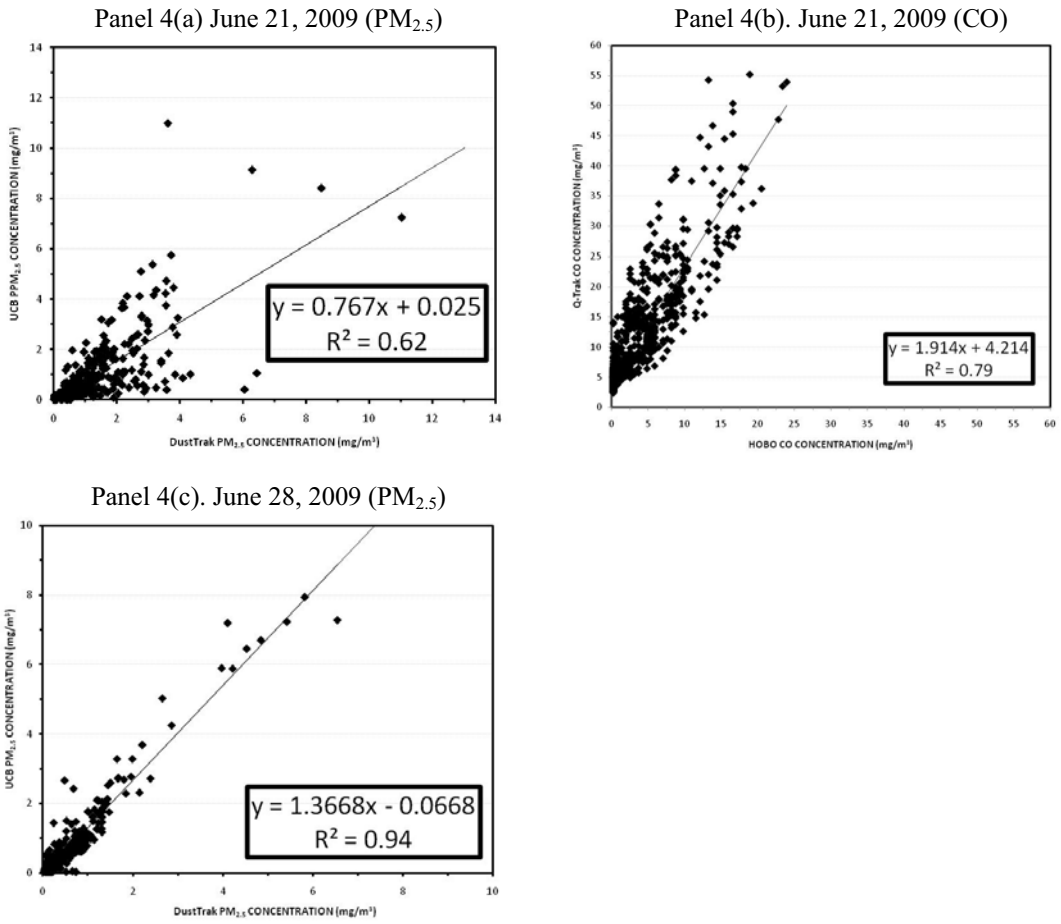


Fig. 4. Correlation between $PM_{2.5}$-measuring instruments, UCB and DustTrak [Panel (a) for June 21, 2009 in the BCSIR improved stove kitchen and Panel (c) for June 28, 2009 in the unimproved mud stove kitchen]. All are adjusted with respective gravimetric standards and corrected for relative humidity. The scatter plots in Figs. 4(a) and (c) reflect the correlation between the UCB and DustTrak as seen in Figs. 3(a) and (b), respectively. If the high points were removed from Fig. 4(a), the new linear equation changes to y = 1.167x + 0.038 with an r^2 = 0.64 which is higher than the current r^2 = 0.62. Panel (b) shows the correlation between CO-measuring instruments, Q-TRAK Plus and HOBO for June 21, 2009 in the BCSIR improved stove kitchen.

Table 3. Descriptive statistics of $PM_{2.5}$ and CO concentration by stove status.

	$PM_{2.5}$ (mg/m^3)		CO (mg/m^3)	
	Mud Stove	BCSIR Stove	Mud Stove	BCSIR Stove
Mean	1.92	1.38	4.90	2.97
Median	1.77	0.73	2.54	1.96
Standard Deviation	1.59	1.27	6.29	2.29
Standard Error	0.37	0.29	1.37	0.49
Minimum	0.22	0.11	0.23	0.28
Maximum	5.85	4.22	25.88	7.13
Range	5.63	4.11	25.65	6.85
Count, N	20	19	21	22

BCSIR improved stove kitchens. If the high point in Fig. 5(a) were removed the r^2 decreases to 0.31, but the fit of the line remains the same (slope 0.61 and intercept 0.23). Households with no simultaneous $PM_{2.5}$ and CO measurements were excluded from this analysis. Five outliers were identified and excluded from the analysis when survey questionnaires confirmed that no cooking took place in the kitchen during the sampling period or when an open fire stove was used in conjunction with the BCSIR improved stove.

A lower r^2 tended to occur for households that were located near each other since neighborhood smoke penetrated from one household into another. During the non-cooking periods, CO concentrations reach the instrumental detection limit of 0.2 ppm and stay constant. $PM_{2.5}$ concentrations

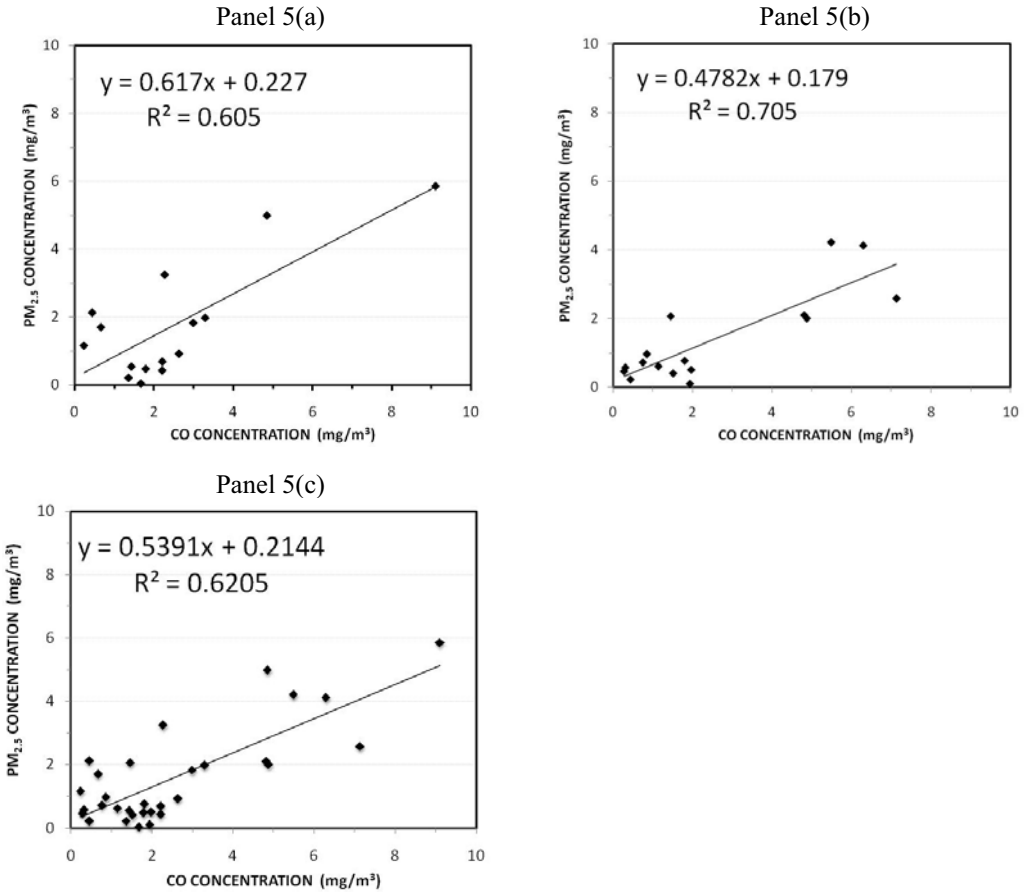


Fig. 5. Correlation between $PM_{2.5}$ and CO in households with (a) the unimproved mud stove (R^2 = 0.61, N = 15), (b) the BCSIR improved stove (R^2 = 0.71, N = 16), and (c) all household with improved and un-improved stoves (R^2 = 0.62, N = 31).

fluctuate between 0.01 and 0.09 mg/m^3 further lowering our correlation coefficient (R^2). Nevertheless, high regression coefficient, r^2, between $PM_{2.5}$ and CO concentrations demonstrates their proportional relationship with each other. Concentrations of CO and $PM_{2.5}$ peak at the same time when cook stoves are typically expected to be used (breakfast, lunch, and dinner). Past studies using solid fuel in biomass burning stoves in Guatemala quantified $PM_{2.5}$ concentrations from the CO concentration values and supports this evidence that $PM_{2.5}$ and CO are co-emitted (Naeher *et al.*, 2001; Northcross *et al.*, 2010). Calculations from a 48 h study in the highlands of Guatemala reported a correlation coefficient, r^2, of 0.76 between CO and $PM_{2.5}$ (Northcross *et al.*, 2010). Naeher *et al.* (2001) reported a correlation, R, of 0.92 (r^2 = 0.84) at a nearby location in Guatemala. Siddiqui *et al.* (2008) in Pakistan, and Park and Lee (2003) in Costa Rica found Spearman correlation coefficient between CO and $PM_{2.5}$ to be 0.72 and 0.71, respectively, as well. However, Ezzati *et al.* (2000) in Kenya found the distribution of PM_{10} relative to CO to be skewed and PM_{10} and CO to peak asynchronously. In the present study, we observe the $PM_{2.5}$ peaks coincide with CO peaks showing PM is at its highest when CO peaks. However, the variation patterns of PM do not match those of CO (see Fig. 3(c)). PM concentrations rise and fall sharply while concentrations of CO show a gradual increase and decrease around the peaks of CO. Depending on the cooking method, PM emissions can vary significantly (Huboyo *et al.*, 2011). Ezzati *et al.* (2000) also observed this trend of sharper fluctuations of PM_{10} with respect to CO in Kenya, explaining his wide range of correlation (r^2 = 0.14 – 0.91) between the two parameters. Because of the differences in emissions during flaming and smoldering of wood fuel, the ratio between CO to $PM_{2.5}$ varies during the burn cycle and can vary between 0.3 and 45. To address the fluctuating CO to PM ratio during the burn cycle of wood fires, the CO to $PM_{2.5}$ correlation was calculated using the 24 hours daily mean concentrations for both CO and $PM_{2.5}$ by stove status as displayed in Eqs. (1) and (2) below. Eq. (1), with 95% confidence interval of the regression coefficient and intercept expressed in parentheses, represents the linear regression of measured concentrations from kitchens using the unimproved mud stoves (N = 15). Eq. (2) represents data from the kitchens using the BCSIR improved stoves (N = 16). Eq. (3) encompasses all data used in Eqs. (1) and (I2), as shown in Fig. 5(c).

$$PM_{2.5}\ (mg/m^3) = 0.62\ (0.32, 0.92) \times CO\ (mg/m^3) + 0.23\ (0.05, 0.48),\ R^2 = 0.61,\ N = 15 \quad (1)$$

$$PM_{2.5}\ (mg/m^3) = 0.48\ (0.30, 0.66) \times CO\ (mg/m^3) + 0.18\ (0.01, 0.39),\ R^2 = 0.71,\ N = 16 \quad (2)$$

$PM_{2.5}$ (mg/m^3) = 0.54 (0.38, 0.70) × CO (mg/m^3) + 0.21 (0.02, 0.35), R^2 = 0.62, N = 31 (3)

The regression coefficients for Eqs. (1–3) in the present study are slightly lower than the regression coefficients found by Northcross *et al.* (2010). In the present study, the $PM_{2.5}$ concentrations are higher compared to the corresponding CO concentrations which is different for this same relationship observed by Northcross *et al.* (2010). This difference may be due to differences in the stove, fuel, and combustion properties as well as the cooking practice between the two studies. In Bangladesh, fuel tends to have higher moisture content leading to higher particulate emissions than the fuel used in Guatemala. The cooking practice varies between the two regions (Asaduzzaman *et al.*, 2009). Moreover, both type of stoves in our study produced high concentrations of $PM_{2.5}$, limiting the correlation to represent only measurements in the upper range of $PM_{2.5}$ concentrations that was not the case for Northcross *et al.* (2010) where both CO and $PM_{2.5}$ concentrations in kitchens with Improved *Plancha* stoves were significantly less than the corresponding concentrations in kitchens with open-fire stoves. The CO to PM regression coefficient found by Ezzati *et al.* (2000) is low which can be explained since their study used multiple fuels and stove types. Our higher r^2 value resulted from only two fuel sources (wood and "Golden", with wood being the primary fuel) and two particular stove types (i.e., unimproved mud stoves and the BCSIR improved stoves). Ezzati *et al.* (2000) also concluded the linear relationship between CO and PM was dependent on the type of fuel and stove. Eq. (3) removes the dependence of stove type as it can be used to predict $PM_{2.5}$ concentration emitted from either the unimproved mud stoves and the BCSIR improved stoves. The fuel source, then, becomes the sole factor influencing emitted PM concentrations. Therefore, Eq. (3) is effective, when there is a single combustion source (Northcross *et al.*, 2010).

These findings aims to allow future low-funded studies to take advantage of this strong correlation between CO and $PM_{2.5}$ to further determine the air quality of indoor air pollution. Traditional PM measurement devices are costly, invasive, logistically complicated, and impossible to use on children and infants (Northcross *et al.*, 2010). However, CO measurements can be made with inexpensive electro-chemical monitors and small diffusion tubes. These instruments are relatively easy to set up and highly affordable.

Neighborhood Air Pollution

As noted above, neighborhood $PM_{2.5}$ and CO were measured by deploying a single gravimetric device and a single HOBO on a daily basis to collect eight 24-hr long samples outside of each house that were part of the intensive monitoring. Mean concentrations of $PM_{2.5}$ and CO were 0.40 ± 0.14 mg/m^3 (range 0.19 – 0.58 mg/m^3) and 1.1 ± 0.40 mg/m^3 (0.35 – 1.5 mg/m^3), respectively. The $PM_{2.5}$ concentrations were high providing an indication of the high outdoor PM air pollution existing in the immediate vicinity of these households. The study area was a tightly packed slum area where hundreds of homes are clustered together in a small area with minimal to no space between adjacent homes. Because of the level of poverty, building homes in urban slum areas are common in Bangladesh, South Asia, and in other parts of developing Asia (Bhaskar and Mehta, 2010). Our outdoor measurements indicated significant air pollution exists in such neighborhoods. Visual observation during field sampling indicated smoke clouds above and around homes, especially during the early and late morning hours when breakfast and lunch were being prepared and the outside temperature was still cool. This neighborhood effect contributed to the high indoor air pollution in many of these homes having the BCSIR improved stove. Smith (2002) also noted that because of the close proximity of the households, neighborhood air pollution causes elevated levels of carbon monoxide and fine particulate matter even when improved stoves are used.

Contribution of biomass to $PM_{2.5}$ in outdoor air in the Indian Subcontinent can be estimated by conducting source apportionment modeling. Using Chemical Mass Balance (CMB) source apportionment in a background site in Chandigarh, India, Chowdhury *et al.* (2007b) estimated 8% of the $PM_{2.5}$ in the outdoor air to be from biomass sources. In the Maldive Islands, a significant amount of potassium, a biomass marker, was quantified suggesting the importance of biomass combustion to the outdoor $PM_{2.5}$ concentration. In Bangladesh, Begum *et al.* (2004) found biomass combustion to constitute approximately 50% of the $PM_{2.5}$ in Rajshahi, a location not too far from Saidpur, our study location. Dasgupta *et al.* (2006) also concluded that outdoor pollution significantly affects indoor air quality because houses are built in clusters and suggested using taller stacks which would permit the exhaust to be diluted as a short-term solution. However, long term solution lies in shifting the entire community or neighborhood to cleaner burning stoves and modern fuels instead of installing improved stoves in a select few.

Carbonaceous Species and Trace Elements Concentrations of Fine Particles

Table 4 presents the concentrations of all chemical species measured during the intensive monitoring. Concentrations of organic carbon (OC) and elemental carbon (EC) were 0.21 ± 0.02 mg/m^3 and 0.18 ± 0.01 mg/m^3 for unimproved mud stoves and 0.12 ± 0.01 mg/m^3 and 0.10 ± 0.01 mg/m^3 for the BCSIR improved stoves, respectively. Also, the ratios were identical for both types of stoves: 0.84 for EC/OC ratio, 0.54 for OC/TC ratio and 0.46 for EC/TC ratio. Begum *et al.* (2009) measured black barbon (BC) and OC in 5 households in rural Savar, Dhaka District, Bangladesh and reported 27–33% BC and 63–77% OC in $PM_{2.5}$ with a BC to OC ratio of 0.43 which is lower than our ratio. Similar studies in rural China report that improved stoves or improved fuel serve to decrease organic and elemental carbon of up to 61% and 98%, respectively (Zhi *et al.*, 2009). Furthermore, elemental carbon/organic carbon ratios appear to be more consistent in the improved stoves (0.82), while this ratio for the unimproved stoves ranges from 1.0 to 0.68 suggesting a variation in the burn cycle and changes in the efficiency of the stoves since a higher EC/OC ratio

suggests more soot being released in the air.

Table 4 also shows the concentrations of trace elements. These values are averaged of four samples for each stove status. Concentrations of Cl, Cu, Fe, K, Na and S are higher than 1 μg/m^3. Potassium has been suggested as a marker for wood combustion and our results indicate K to be present at high concentrations in the kitchen since the predominant fuel is wood. Metals with multiple toxic effects such as As, Cd, Cr, Pb, Hg, Ni have also been detected in these samples. The health effects associated with some of these inorganic compounds as well as organic compounds have been presented in details in Naeher *et al.* (2007). A chemical mass balance of the fine particulate matter was conducted by summing all identified chemical species in

Table 4. $PM_{2.5}$ chemical composition in the unimproved mud stove kitchen, BCSIR improved stove kitchen, and in the neighborhood outdoor air. Concentrations are expressed in μg/m^3 units. Bold numbers indicate greater than 2X Abs Err. ND = no data available since the quartz filter was not used in this sample.

Species		Unimproved Mud Stove		BCSIR Improved Stove		Neighborhood Pollution	
		Mean	Abs Err	Mean	Abs Err	Mean	Abs Err
1	TC	**387**	27	**222**	23	ND	ND
2	EC	**176**	13	**152**	11	ND	ND
3	OC	**210**	15	**182**	12	ND	ND
4	Ag	0.029	0.033	0.021	0.036	0.000	0.000
5	Al	**0.141**	0.040	**0.123**	0.041	0.010	0.019
6	As	0.010	0.017	0.015	0.017	0.009	0.010
7	Ba	0.089	0.423	0.062	0.441	0.000	0.342
8	Bi	0.006	0.009	0.013	0.009	0.007	0.006
9	Br	**0.070**	0.014	**0.156**	0.011	**0.071**	0.010
10	Ca	**0.285**	0.018	**0.263**	0.023	**0.255**	0.019
11	Cd	0.009	0.047	0.014	0.034	0.000	0.019
12	Ce	0.061	0.341	0.146	0.418	0.478	0.578
13	Cl	**31.927**	0.086	35.005	0.11	**3.769**	0.006
14	Co	0.038	0.059	0.039	0.062	0.037	0.042
15	Cr	0.005	0.007	0.004	0.007	0.005	0.005
16	Cs	0.000	0.336	0.046	0.289	0.105	0.172
17	Cu	**1.106**	0.105	**1.213**	0.108	**1.015**	0.075
18	Fe	**2.901**	0.192	**2.908**	0.199	**2.844**	0.135
19	Ga	0.007	0.028	0.004	0.029	0.000	0.019
20	Ge	0.000	0.001	0.000	0.001	0.000	0.001
21	Hg	0.007	0.012	0.000	0.001	0.012	0.013
22	I	0.000	0.295	0.041	0.271	0.000	0.208
23	K	**13.507**	0.110	**10.857**	0.096	**5.768**	0.040
24	La	0.000	1.138	0.217	0.982	0.320	0.638
25	Mg	0.080	0.046	**0.151**	0.074	0.030	0.020
26	Mn	0.018	0.010	0.013	0.01	0.009	0.007
27	Mo	0.307	0.268	0.378	0.237	0.242	0.208
28	Na	**3.840**	0.941	**2.158**	0.934	**0.683**	0.331
29	Ni	**0.469**	0.073	**0.508**	0.075	**0.501**	0.051
30	P	**0.107**	0.007	**0.109**	0.007	**0.043**	0.002
31	Pb	**0.089**	0.038	**0.090**	0.038	**0.096**	0.026
32	Rb	**0.060**	0.012	**0.086**	0.011	**0.030**	0.010
33	S	**4.336**	0.016	**3.944**	0.016	**3.036**	0.006
34	Sb	0.000	0.001	0.000	0.001	0.000	0.001
35	Sc	0.003	0.007	0.002	0.007	0.006	0.005
36	Se	0.010	0.006	0.005	0.007	0.000	0.000
37	Si	**0.117**	0.025	**0.070**	0.026	**0.064**	0.013
38	Sm	0.004	0.011	0.002	0.011	0.000	0.007
39	Sn	0.048	0.099	0.021	0.101	0.072	0.069
40	Sr	**0.043**	0.017	**0.040**	0.017	**0.042**	0.012
41	Te	0.027	0.138	0.000	0.164	0.000	0.111
42	Ti	0.007	0.007	0.005	0.008	0.001	0.005
43	V	0.005	0.006	0.003	0.006	0.003	0.004
44	W	0.012	0.047	0.008	0.048	0.022	0.032
45	Zn	**0.129**	0.033	**0.158**	0.035	**0.164**	0.025

the two types of kitchens during the intensive monitoring (see Fig. 6). OC was converted to organic matter (OM) as outlined in Chowdhury *et al.* (2001). Particulate organic matter was calculated using a conversion factor of average organic molecular weight (for molecules O, N, H) per carbon weight to represent the organic matter within $PM_{2.5}$. This factor was 1.7 (Turpin and Lim, 2001). Of the identified chemical species, 29–30% is EC in nature and 59–60% is OM in nature. Chlorine and potassium are 5–7% and 2%, respectively, of the identified species. These % contributions are within the range reported by Calvo *et al.* (2011) for biomass combustion.

Estimating Personal Exposure and Health Effects

During our study period, to obtain a general understanding of the health effects of the cooks (women in their reproductive age) who spend the most time in the kitchen, we calculated their inhaled dose of $PM_{2.5}$ and CO via applying to the dose Eq. (4) below the inhalation rate of approximately 18 m^3/24 hr (Smith and Peel, 2010), the average exposure concentration from each stove type, and the respective average exposure duration.

Dose = inhalation rate (m^3/hr) × avg. conc. (mg/m^3) × avg. exposure duration (hr) (4)

The calculated average daily dose exposure to $PM_{2.5}$ of cooks with the BCSIR improved stoves was 4.4 mg and to CO was 9.4 mg. Cooks using the unimproved mud stoves were exposed to about 5.8 mg of $PM_{2.5}$ and 14 mg of CO during their average cooking time in one day. Smith and Peel (2010) suggested an elevated relative risk (RR) of cardiovascular disease (CVD) linking to the concentration of $PM_{2.5}$ inhaled from indoor biomass burning. This theory calls for further evidential support by epidemiological study on cardiovascular risks of indoor air pollution. However, there exists support for influences of $PM_{2.5}$ on blood pressure (McCracken *et al.*, 2007). If Smith and Peel's theory is accepted, then the RR of CVD for cooks using the BCSIR stove is approximately 1.5. For cooks using the unimproved mud stove, the RR is higher than 1.5. Although the two stove types do not provide a significant difference in relative risk of CVD, it should be noted that the risk for CVD from inhaling indoor $PM_{2.5}$ is 50% higher than normal. The risk of developing chronic obstructive pulmonary disease (COPD) associated with the use of biomass fuel is increased in adult women from long-term exposure to smoke from unventilated stoves (Smith and Mehta, 2003). In addition, there is emerging evidence which suggests that indoor air pollution from biomass smoke also increase the risk of tuberculosis, cataract, and upper airway cancer (WHO, 2002).

Carbon monoxide diminishes health conditions by competing with oxygen for binding to hemoglobin (Hb). The attraction of Hb to CO is approximately 200 times stronger than that for oxygen (Rosenthal *et al.*, 2006). Blood COHb saturation in an average nonsmoker ranges from 1–3% due to varied endogenous production and exposure to 0.1 ppm CO in ambient air (USEPA, 2000; Kao *et al.*, 2004). Mild symptoms, such as nausea, vomiting, dizziness, blurred vision and headache, may occur in individuals with COHb level less than 15–20%; COHb level greater than 60–70% is increasing fatal with symptoms, such as cardiac arrest and coma (Kao *et al.*, 2004).

Lee *et al.* (1994) conducted a study of CO-COHb relationships where participants are exposed to 9 ppm of CO for time intervals of 1 hour and 8 hours, and compared to baseline values at 0 hour. The linear equation they found for relationship between breath CO concentration and blood COHb saturation is X = 0.01 + 0.26(Y) for the mean, where X is the percent of COHb in blood and Y is the breath CO concentration (ppm). The mean CO concentrations from 1 and 8 hours of exposure in our study yields COHb saturation of 11% and 3.8% in control households and 8.8% and 2.5% in intervention households over the respective time intervals when using the aforementioned equation. These percentage values are applicable to cooks living in the households where CO exposure exceeds the median concentrations in households under the extensive monitoring. WHO guidelines permit COHb saturation in the blood below 2.5%; however, our calculations indicate that 50% of our cooks in both the control and intervention homes exceed the WHO guidelines. It can be inferred that these cooks may suffer from the above listed mild symptoms associated with carbon monoxide poisoning during their daily cooking activities.

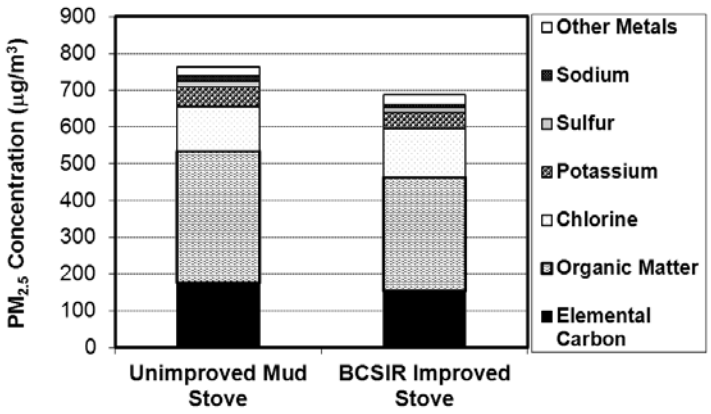


Fig. 6. Comparison of $PM_{2.5}$ mass balance from all identified chemical species in the two types of kitchens during the intensive monitoring.

Limitations

There are limitations in the research that potentially limited the quality of the results. Some HHs in the intervention group elected to use their unimproved mud stoves to supplement the cooking done with the BCSIR improved stoves provided. Although, the fuel source for both types of cookstoves was the same, the concentration of pollutants emitted by the unimproved stove contributed the measurements of CO and PM concentration. Through observation, it was noted that the cook(s) tend to leave open one of the two pot stances available in the improved stove. This opening allowed pollutant-carrying smoke to escape into the kitchen, and contributing to higher pollutant concentrations inside the house. This design issue requires stove engineering specialists to consider future improvements in the BCSIR improved stoves. Aside from those directly related to cooking, additional combustion sources were present, such as: secondary lighting of kerosene, mosquito coils, and smoke from nearby kitchens (see Table 1). All of these contributed to higher level of CO and PM than would be otherwise. The subjective answers received from the cooks further add to our limitations in this research. Answers for the time spent cooking were not measured, and were based on the memory of the cook(s)'s activities in the kitchen the day before.

CONCLUSIONS

This pilot project highlights the reliance on biomass fuels and the factors contributing to air pollution inside homes in Bangladesh. Cooking with the BCSIR improved stove instead of the unimproved mud stove lowered CO and $PM_{2.5}$ concentrations by 23% and 59%, respectively. The predominant chemical species are elemental carbon, organic matter, chlorine, and potassium. The emitted $PM_{2.5}$ chemical composition was 59–60% organic matter and 29–30% elemental carbon. This study was the first to measure and analyze the relationship between CO and $PM_{2.5}$ within the rural region of Bangladesh. Moreover, this research was also the first to evaluate findings of ultrafine particles in an indoor setting suggesting emissions from biomass cook stoves may penetrate very deep into the respiratory and cardiovascular system. From this IAP work, we demonstrated the practicality and feasibility of performing low-funded indoor air pollution study in rural Bangladesh.

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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Supplemental Methods

S.1 Stove Characteristics

The BCST stoves are improved stoves with an iron grate and a chimney to conduct the flue gases outside the house through a hole in the roof. This type of stove was available in three model types: three-pot, two-pot and single-pot improved stoves. The majority of the project households in our study used the two-pot BCST improved stoves. A Standard Water Boiling Test (WBT) was performed by Appropriate Rural Technology Inc. (ARTI), an Indian NGO, following the Aprovecho and UC Berkeley method for this test (MacCarty *et al.*, 2008). When starting cold, the 1-pot traditional mud stove took 20 min to boil the same amount of water as opposed to the BCST stoves (13 min and 18 min for the 1-pot and 2-pot stoves, respectively). Whereas, when starting hot, the 1-pot traditional mud stove took 17 min, as opposed to 10 min and 13 min for the 1-pot and 2-pot BCST stoves, respectively. Fig. (2) shows the improved and traditional stoves with design parameters as well as views of the primary and secondary fuels used in this study.

Thermal efficiencies reported by ARTI were 0.25 for the traditional one-pot stove and ranged between 0.11 and 0.2 for the BCST stoves. Also, specific fuel consumption, which is the fuel required to boil 2.5 liters of water was measured. Approximately 100 g/L was used for the traditional stove as compared to 0.18 g/L for the BCST stoves. These parameters helped in quantifying the performance of the BCST improved stove.

S.2 Instrument Protocol

$PM_{2.5}$ was measured gravimetrically using the two co-located SKC AirCheck XR5000 programmable pumps operated by internal Ni-Cd battery using a BGI SCC1.062 Triplex cyclone. The target flow was 1.5 LPM to achieve a 50% cut point (50% penetration) for $PM_{2.5.}$ Two flow rates (initial and final) were measured with a field rotameter calibrated against a Gilibrator (electronic bubble flowmeter) in the lab. One of the two co-located gravimetric devices used a 37-mm TEFLO PTFE membrane filter with PMP ring (SKC West, part # '225-1709 pore size of 1 micron), while the other used a 37-mm Whatman QMA quartz filter (BGI USA, part 1851037) as the particle collection media. The same filter lot was used for each of the filter media. As described in section 2.6, the Teflon filters were analyzed for gravimetric mass and 42 trace elements, whereas the quartz filters were analyzed for elemental and organic carbon. The Teflon filters were pre-weighed and post-weighed with a 6-place Sartorius microbalance at San Diego Air Pollution Control District (SD APCD). The weighing room was controlled for temperature (T) and relative humidity (RH). Measurements for T and RH were reported during the weighing period. Static electricity was discharged before each weighing by passing each side of the filter near a polonium 210 alpha-radiation source for a few seconds. Filter blanks were weighed after every 10 sample filter weights. Each filter was weighed at least twice until the mass difference between the repeated weighing was equal to or less than 5 μg.

Before each use, the DustTraks and the P-TRAKs were zeroed according to the manufacturers' instructions using a HEPA filter, their flowrates checked, and the time synchronized with the laptop's internal clock. One minute interval logging consisting of averaging second-by-second sampling was set. The alcohol wick of the P-TRAKs were charged every 7 hours in the field. During instrumental launching, the internal clock on the UCB was synchronized by the Monitor Manager© software to the clock on the field laptop, which was set to atomic time by internet once per week. Prior to going to the field, the supervisor's wristwatch was synchronized to the time on the same computer so that field notes could be easily related to the UCB data. Following the protocols developed for the project, the UCBs were zeroed before and after every monitoring event by placement inside a particle-free sealed plastic bag for approximately 30 minutes each time. All HOBOs and UCBs were calibrated at SDSU and in Dhaka before the start of the field work. The HOBOs and Q-TRAKs were calibrated with span gases with known concentrations of CO and the UCBs were cleaned and calibrated against gravimetric measurement with the help of a DustTrak. Temperature and humidity were measured by the UCB, HOBO T-RH Monitor (H08-003-02 Onset Corp.) and the Q-TRAK during the intensive monitoring and by both the UCB P3 and HOBO T-RH Monitor (H08-003-02 Onset Corp.) during the extensive monitoring.

Strict guidelines were applied in placing all monitoring equipment in the kitchens. Instruments were placed on the wall approximately 100 cm from the center of the combustion zone of the

cooking stove. This distance was measured as the shortest, horizontal line (i.e. parallel to the floor). The instruments were placed 145 cm above the floor defined as the level directly below the monitor. This height relates to the approximate breathing zone of a standing woman. Finally, the equipment were located at least 150 cm away (horizontally) from doors and windows, where possible.

Supplemental References

MacCarty, N., Ogle, D., Still, D., Bond, T. and Roden, C. (2008). A Laboratory Comparison of the Global Warming Impact of Five Major Types of Biomass Cooking Stoves. *Energy for Sustainable Development* 12(2): 5-14.

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WHO (2006). WHO Air Quality Guidelines: Global Update for 2005. Copenhagen: World Health Organization Regional Office for Europe.

Supplemental Tables

SI Table 1. Frequency distribution of households exceeding WHO guidleines for CO and PM. WHO ambient air quality guidelines for CO are based on mathematical estimates to prevent carboxyhemoglobin levels in the blood from exceeding 2.5%.

	WHO Guideline (ppm for CO); (ug/m^3 for PM)	Quantity of Household Exceeded (number and Percentage)
15-min CO average	90[a]	6; 15.38%
1-hr CO average	25[a]	12; 30.77%
8-hr CO average	10[a]	13; 33.33%
24-hr PM2.5	25[b]	40; 100%
24-hr PM10	50[b]	ND

a WHO, 2004

b WHO, 2005; WHO, 2006

Supplemental Figures

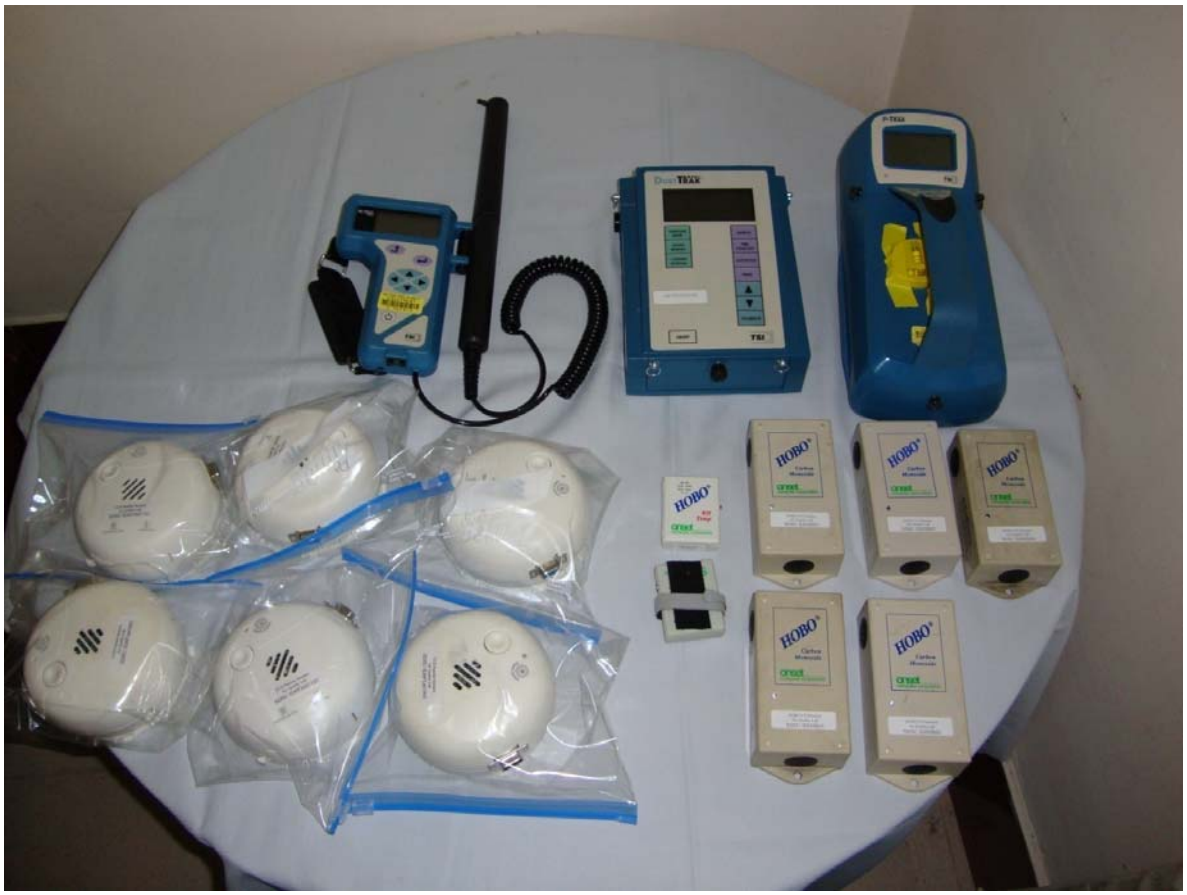

Fig. (S1). Photos of the instruments deployed in the field.