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Evaluation of exposure reduction to indoor air pollution in stove intervention projects in Peru by urinary biomonitoring of polycyclic aromatic hydrocarbon metabolites

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ABSTRACT

Burning biomass fuels such as wood on indoor open-pit stoves is common in developing regions. In such settings, exposure to harmful combustion products such as fine particulate matter ($PM_{2.5}$), carbon monoxide (CO) and polycyclic aromatic hydrocarbons (PAHs) is of concern. We aimed to investigate if the replacement of open pit stoves by improved stoves equipped with a chimney would significantly reduce exposure to PAHs, $PM_{2.5}$ and CO. Two stove projects were evaluated in Peru. Program A was part of the Juntos National Program in which households built their own stoves using materials provided. In Program B, Barrick Gold Corporation hired a company to produce and install the stoves locally. A total of 30 and 27 homes participated in Program A and B, respectively. We collected personal and kitchen air samples, as well as morning urine samples from women tasked with cooking in the households before and after the installation of the improved stoves. Median levels of $PM_{2.5}$ and CO were significantly reduced in kitchen and personal air samples by 47–74% after the installation of the new stoves, while the median reduction of 10 urinary hydroxylate PAH metabolites (OH-PAHs) was 19%–52%. The observed OH-PAH concentration in this study was comparable or higher than the 95th percentile of the general U.S. population, even after the stove intervention, indicating a high overall exposure in this population.

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1. Introduction

Globally, approximately 3 billion people (52% of the population) rely on biomass, such as wood, coal, charcoal, and crop residues, as their primary source of domestic energy (Rehfuess et al., 2006). Exposure to indoor air pollution (IAP) from biofuels has been found to be one cause of respiratory diseases in developing counties (Ezzati and Kammen, 2001b; Mehta et al., 2002). In addition, a growing number of studies have linked IAP from solid fuel usage to other health conditions such as low birth weight and stillbirth (Boy et al., 2002; Pope et al., 2010). Indoor smoke from solid fuels was suggested to be the fourth leading risk factor (after underweight, unsafe sex, and poor water sanitation and hygiene) of disease burden in high-mortality developing regions such as India, killing 1.6 million people worldwide, and contributing to 39 million burden of disease (Ezzati et al., 2002). Acute respiratory infection, one of the leading diseases that accounts for more than 6% of worldwide morbidity and mortality, has been found to possess positive exposure–response relations with domestic biomass burnings (Ezzati and Kammen, 2001a).

In developing countries where solid fuel usage is the most prominent, biomass fuels are often burnt in open fire pits or in poorly constructed stoves in rooms with no or poor ventilation, contributing to high levels of harmful incomplete combustion products inside the house/kitchen (Naeher et al., 2007). Poor combustion efficiency results in formation and emission of a large number of harmful pollutants including fine particles ($PM_{2.5}$), carbon monoxide (CO), nitrogen oxides, formaldehyde, and polycyclic aromatic hydrocarbons (PAHs) such as benzo(a)pyrene, a known carcinogen. Hazardous compounds have also been found in solid residues, such as ashes from wood combustion (Oehme and Muller, 1995; Wunderli et al., 2000) and soot from wood stoves (Wu et al., 2002).

Women and children often bear disproportionally more burden of IAP-related health effects in developing countries, since they generally

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Abbreviations: PAH, polycyclic aromatic hydrocarbons; OH-PAH, hydroxylated polycyclic aromatic hydrocarbon metabolites; NHANES, National Health and Nutritional Examination Survey; GC/HRMS, gas chromatography/high-resolution mass spectrometry; $PM_{2.5}$, fine particulate matters with aerodynamic diameters less than 2.5 μm; IAP, indoor air pollution.

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spend more time at home and in the kitchen (Manuel, 2003; Smith et al., 2004). More than 500,000 of the deaths of 1.3 million women globally from chronic obstructive pulmonary disease (COPD) were attributable to IAP, compared to only 171,000 of 1.4 million COPD deaths linked to IAP among men (Smith et al., 2004). Infants and children are especially susceptible to biomass fuel-related IAP because they spend a great deal of time near their mothers, and they inhale a greater volume of air per unit body mass than an adult. In addition, their airways and immune systems are still developing, making them more susceptible to adverse health outcomes. Reports have stated that IAP is responsible for over 900,000 annual deaths resulting from acute lower respiratory infection in children <5 years of age, which corresponds to 56% of all IAP-attributable deaths (Rehfuess et al., 2006; Smith et al., 2004).

Given that most biomass fuels are readily available and relatively cheap or free, they will likely remain the major domestic energy source in many parts of the world, especially in economically developing regions. As a result, the most feasible and cost-effective measure, especially in rural areas, to reduce IAP and human exposure to IAP may reside in the installation and usage of improved stoves that reduce the emission of harmful combustion products and/or reduce human contact to such hazards by diverting the exhausts to the outside of the home. In urban areas, other solutions may be necessary including the use of cleaner-burning fuels to reduce exposure resulting from the close vicinity of homes. Recently, stove intervention programs or projects have been implemented or are in planning in a number of countries/regions, such as China (Edwards et al., 2007; Zhang and Smith, 2007), India (Venkataraman et al., 2010), Mexico (Romieu et al., 2009; Torres-Dosal et al., 2008), Guatemala (Granderson et al., 2009; Smith-Sivertsen et al., 2009), Honduras (Clark et al., 2009), and Canada (Allen et al., 2009). Most noteworthy, Global Alliance for Clean Cookstoves, an initiative led by the United Nations Foundation that incorporated global partnership including a number of U.S. federal government agencies, other country governments, private corporations, and non-profit organizations, was launched on September 21, 2010 (Glocal Alliance for Clean Stoves, 2010). The Alliance's overarching goal, "100 by 20," calls for 100 million households to adopt clean and efficient stoves and fuels by 2020, which will further the global effort on this front.

In Peru, 36.9% of homes (over 90% in rural areas) use solid fuels for cooking and heating, based on Peruvian census (INEI, 2007). In June 2009, the Peruvian government began a nationwide intervention program to install 500,000 improved stoves by December 2011, set to comply with the United Nations Millennium Development Objectives (Rehfuess et al., 2006) for Energy and Poverty Alleviation targeting poor households, mostly at high altitudes, to improve health and reduce poverty. Meanwhile, Barrick Gold Corporation in Peru has reached out to their local communities and begun providing improved stoves as well. We conducted an intervention study to evaluate the effectiveness of the improved stoves in two separate programs through air monitoring and biomonitoring. We measured $PM_{2.5}$ and CO in kitchen and personal air samples in 57 participating households before and after installation of new stoves with chimney. In addition, we measured 10 urinary hydroxylated PAHs (OH-PAHs), biomarkers of PAH exposure, in morning urine samples collected from female participants before and after the installation of the improved stoves.

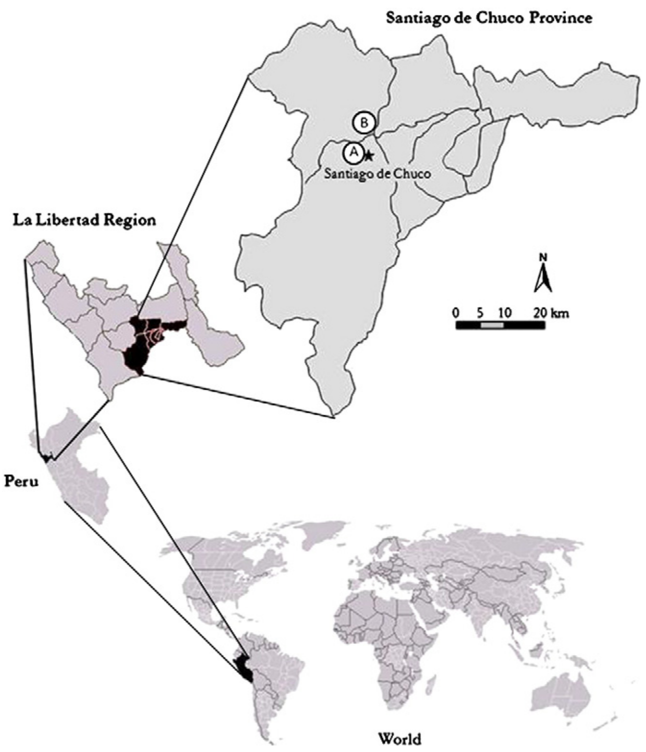

Fig. 1. Map of Santiago de Chuco Province in the La Libertad Region of Peru. Program A was implemented in Huayatan (A), and Program B was implemented in Chaguin and Cachulla Baja.

2. Material and methods

2.1. Study design

This investigation took place in three locations within the Santiago de Chuco Province in north central Peru (Fig. 1). All homes in these sampling locations use wood for their cooking and heating purposes. The first of two stove intervention programs, hereafter referred to as "Program A," was a part of the Juntos National Program. Program A was carried out in Huayatan, just outside the city of Santiago de Chuco, from June 2 to August 13, 2008. Huayatan is a small, rural community of no more than 100 homes dispersed throughout rolling hills and rugged terrain. Huayatan has one main dirt road, with very little motor vehicle traffic. The stoves in this program were delivered to the homes in three pieces: a 3-pot stove-top, an aluminum chimney, and an aluminum tube to connect the chimney to the stove. These pieces were constructed locally by metalworkers from Santiago de Chuco, and cost approximately 21 US dollar. The stove materials were purchased by the local municipal government, and the participating families built the new stoves in the same room as the previous stove. The second program, Program B, was provided by Barrick Corp. (Lima, Peru). Program B was carried out in two neighboring rural communities, Chaguin and Cachulla Baja, approximately 32 km outside of Santiago de Chuco. These two communities had a combined population of approximately 60 homes. They do not have electricity and are difficult to access by motor vehicles, so vehicle traffic is very limited. The structure of the improved stoves in this project was similar to those in Program A. A local brick mason was contracted to build and install all the stoves. The pre-intervention phase of the study was carried out from June 12 to July 1, 2008, and the post-intervention phase took place September 22–October 3, 2008.

Female subjects were chosen to be part of the study based upon several criteria: they had to 1) be of child-bearing age (18–45 years old) and be the primary cook in the household, 2) use an indoor open fire pit (Fig. 2A) for cooking in a kitchen with at least three and a half full walls and a roof, and 3) not live with a smoker. In Program A, 35 subjects/homes were enrolled; 30 provided urine samples and completed both phases of the study. In Program B, 32 subjects/homes were enrolled, and 27 completed the study. Upon enrollment, each participant filled out a detailed questionnaire, a consent form, and a time activity diary updated every 15 min. In pre-intervention phase, morning voids (50 mL), 48-hour personal and kitchen air samples were taken in all subjects/homes while the original stoves were in use. Post-intervention phase was conducted approximately three weeks after the installation of the new

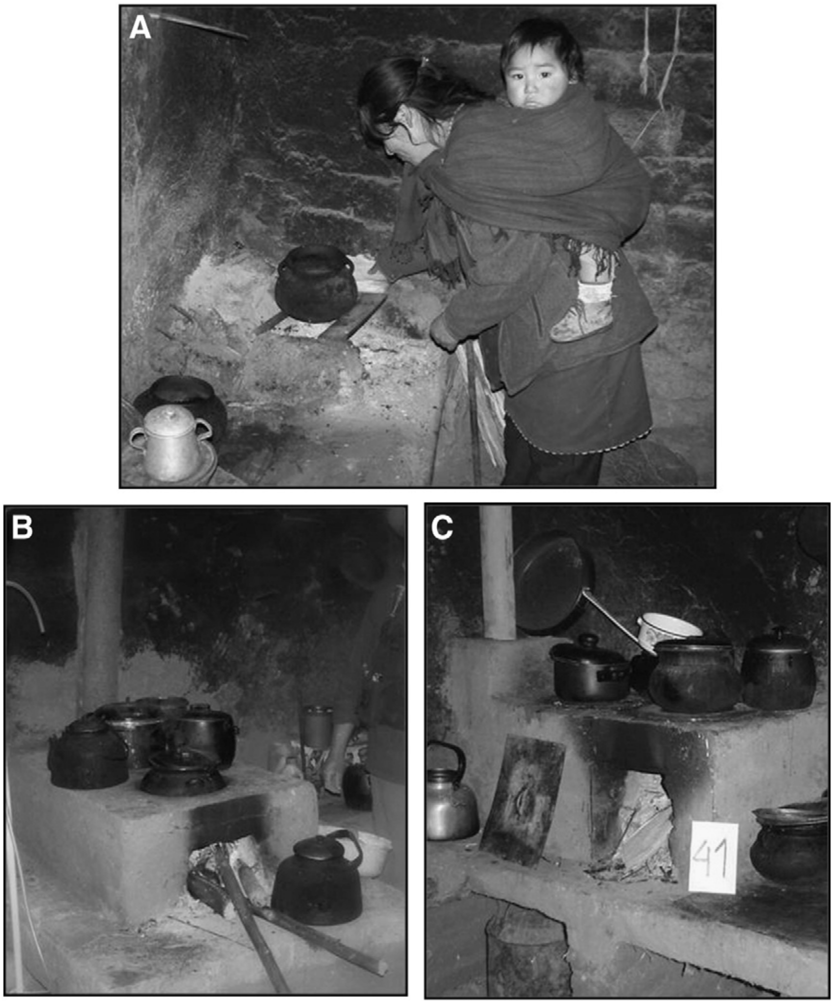

Fig. 2. Examples of indoor open-pit stove before intervention (A), and improved stoves in Program A (B) and in Program B (C).

stoves, during which a second set of questionnaires and time activity diaries was administered. Personal and kitchen air samples and morning voids were also collected post intervention.

2.2. Kitchen and personal air samplings

$PM_{2.5}$ and CO concentrations were monitored in 48-hour personal and kitchen air samples from subjects/homes before the installation of the new stove and three weeks after the new stoves had been in use. A total of 26 subjects/homes in Program A and 18 in Program B completed the air samples in both phases. At the same time, stationary air samplers were also set at a fixed site at the town centers to collect ambient air samples during the two phases of the study. Methods for air sampling and air measurements have been described in detail elsewhere (Fitzgerald, 2009). In brief, the subject wore a vest holding the personal air sampling equipments at or near their breathing zone. Each vest contained a Pac III CO monitor (Draeger Safety Inc., Pittsburgh, PA) and a university sampling pump (SKC Inc, Eighty Four, PA, 1.5 L/min) equipped with particle-size-selective Triplex Cyclones (BGI Inc., Waltham, MA). To collect kitchen air samples, a stationary sampling box containing all necessary equipments was placed within 1 m of the stove in all participating households. A PVC tube was connected to the sampling device and was placed at approximate breathing height (1.5 m). All homes and subjects were sampled for 2 days continuously.

2.3. Urine sampling and analytical method

Morning voids were collected from study participants both before and after the installation of new stoves. Ten metabolites of naphthalene, fluorene, phenanthrene, and pyrene were measured in urine (2 mL). We used a semi-automated liquid–liquid extraction and isotope dilution gas chromatography/high-resolution mass spectrometry (GC/HRMS) method (Li et al., 2006). Urinary creatinine was measured on a Roche Hitachi 912 Chemistry Analyzer (Hitachi Inc., Pleasanton, CA) by use of the Creatinine Plus Assay, as described in Roche's Creatinine Plus Product Application # 03631761003.

2.4. Data and statistical analysis

All urinary OH-PAH concentrations were blank-subtracted, and creatinine adjusted concentrations (ng/g creatinine) were calculated to correct for urine dilution. All $PM_{2.5}$ concentrations were subtracted by the average field blank levels of 0.88 $\mu g/m^3$. Air measurements with sampling duration shorter than 42 h or longer than 54 h were excluded from the analysis. Statistical analyses were performed using the Statistica 7.1 software (StatSoft Inc., Tulsa, OK). Because of the small sample size (30 and 27 urine samples, 26 and 18 air samples for Program A and B, respectively), non-parametric Wilcoxon matched pairs test was used to test the differences between pre- and post-intervention samples, Mann–Whitney *U* test was used to test group differences, and Spearman's rank correlation coefficients were used to evaluate the correlation between urinary OH-PAH concentrations and air pollutant levels. Results are considered statistically significant at $p<0.05$ and marginally significant when the p-value ranged 0.05–0.10.

3. Results

All 10 OH-PAHs were detected well above detection limits in all participating subjects. Table 1 gives the median concentration and quartile range (creatinine adjusted) of the OH-PAH biomarkers in urine collected pre- and post-intervention, stratified by stove types and for all subjects combined. Before the installation of the new stoves, the median concentration of the 10 urinary metabolites in all subjects (n = 57) ranged from 0.4 μg/g creatinine for 4-hydroxyphenanthrene (4-PHEN) to 19.9 μg/g creatinine for 1-naphthol (1-NAP). After the intervention, the urinary levels were significantly reduced for all 10 OH-PAHs, and the median reduction ranged 19% for 3-hydroxyphenanthrene (3-PHEN) to 52% for 1-NAP, cf. Table 1. In Program A (n = 30), the urinary OH-PAH concentrations were reduced by 15% (1-hyroxyphenanthrene or 1-PHEN) to 57% (1-NAP). A statistically significant reduction was observed for 7 out of the 10 biomarkers measured, and a marginally significant reduction was observed for 4-PHEN and 1-PYR (Table 1). In Program B, significant reduction occurred in urinary concentration of 8 metabolites with a median reduction of 13% (3-PHEN) to 38% (3-hydroxyfluorene or 3-FLUO). No significant change in the level of 4-PHEN and 1-hydroxypyrene (1-PYR) was observed in Program B.

Overall, median $PM_{2.5}$ concentration in kitchen area air and personal air at inhalation level before intervention was 181 and 133 μg/m^3 respectively, compared to 77 and 70 μg/m^3 after the installation of the new stoves. The median CO concentrations were 3.5 and 1.2 ppm in kitchen area and personal air pre-intervention, and 0.9 and 0.6 ppm post-intervention (Fitzgerald, 2009). The ambient $PM_{2.5}$ concentrations monitored at stationary sites in the town centers averaged 12.1 (Program A) and 11.6 μg/m^3 (Program B), while the ambient CO levels were 0.25 (Program A) and 0.53 ppm (Program B). The ambient $PM_{2.5}$ and CO level did not change significantly from pre- to post-intervention.

The urinary PAH biomarker levels were significantly correlated with $PM_{2.5}$ and CO in kitchen and personal air in the pre-intervention phases. Spearman's rank correlation coefficients ranged 0.33–0.52 ($p<0.05$) between the summed urinary OH-PAH concentration and $PM_{2.5}$ and CO in kitchen and personal air. In the second phase, significant correlations were still observed, although the strength of the association was reduced (Supplemental Material Table S1).

4. Discussion

In light of the large proportion of the global population that uses domestic solid fuels and the health effects associated with IAP resulting from biomass fuels (Naeher et al., 2007; Rehfuess et al., 2006), stove intervention projects or programs have been conducted in various parts of the world in recent years to reduce human exposure to IAP. In past studies of stove replacement programs, various approaches have been used to evaluate the effectiveness of the improved stoves, including analyzing smoke constituents such as $PM_{2.5}$ and CO in kitchen air and personal air samples (Clark et al., 2010; McCracken et al., 2007), measurement of body functions such as lung function and blood pressure or biomarkers such as blood carboxyhemoglobin (McCracken et al., 2007; Torres-Dosal et al., 2008), and assessing respiratory and other health symptoms through questionnaires (Romieu et al., 2009; Smith-Sivertsen et al., 2009). A commonly used evaluation approach is through measurement of smoke constituents in air. Although this approach produces a good indication of exposure at household levels, the individual exposure can be affected largely by personal behavior patterns and individual physiological differences; hence, the use of a biomarker is preferred to assess the change in exposure. The monitoring of wood smoke-related health outcomes such as respiratory symptoms to evaluate the effectiveness of new cook stoves (Romieu et al., 2009; Smith-Sivertsen et al., 2009) is another approach that requires longer follow-up, on-site infrastructure facilities (access to study subjects and electricity in many cases), and a larger sample size. Therefore, such monitoring is less feasible in the short term to assess reductions of exposure.

We conducted a stove intervention study in Peru to evaluate exposure reduction after replacing wood-burning pits with new stoves equipped with chimneys (Fig. 2). In morning urine samples collected from 57 female subjects, we quantified 10 urinary PAH biomarkers, metabolites of naphthalene, fluorene, phenanthrene, and

Table 1
Median and quartile range of $PM_{2.5}$, CO, and urine OH-PAH concentration (creatinine adjusted, μg/g creatinine) before and after the installation of intervention stoves.

	Abbr.	Pre	Post	% Reduction	Pre	Post	% Reduction	Pre	Post	% Reduction
Urine measurements		All, n = 57			Program A, n = 30			Program B, n = 27		
1-naphthol	1-NAP	19.9 (9.2–25.9)	9.6 (7.8–15.6)	52%***	21.3 (7.9–25.7)	9.2 (6–15.3)	57%***	15 (10.6–27)	10.1 (8–17.5)	33%*
2-naphthol	2-NAP	19.1 (10.8–26)	10 (7.6–15)	48%***	20.3 (9.4–26)	9.9 (6.5–13)	51%***	17.2 (11.9–26.2)	11.4 (8.5–17.1)	34%**
9-OH-fluorene	9-FLUO	3.1 (1.6–4.9)	2.4 (1.5–3.5)	23%**	3.2 (1.5–4.6)	2.2 (1–3.5)	31%*	3 (1.8–5.9)	2.5 (1.8–3.6)	17%*
3-OH-fluorene	3-FLUO	1.2 (0.8–1.9)	0.8 (0.6–1.3)	33%***	1.3 (0.8–2)	0.9 (0.5–1.3)	32%*	1.2 (0.9–1.9)	0.7 (0.6–1.4)	38%**
2-OH-fluorene	2-FLUO	3.7 (2.5–4.6)	2.5 (1.6–3.4)	32%***	3.8 (2.3–4.6)	2.5 (1.4–3.4)	34%*	3.4 (2.6–5.6)	2.3 (1.7–4.2)	32%**
4-OH-phenanthrene	4-PHEN	0.44 (0.22–0.63)	0.3 (0.2–0.5)	30%*	0.5 (0.2–0.6)	0.3 (0.2–0.5)	27%#	0.4 (0.3–0.7)	0.3 (0.2–0.5)	24%
3-OH-phenanthrene	3-PHEN	1.6 (1–2.3)	1.3 (0.7–1.6)	19%***	1.7 (1–2.3)	1.3 (0.7–1.7)	24%*	1.5 (1–2.4)	1.3 (0.9–1.5)	13%*
1-OH-phenanthrene	1-PHEN	1.9 (1.3–3)	1.5 (1.1–2.5)	21%*	1.8 (1.2–3)	1.5 (1.2–2.6)	17%	1.9 (1.3–3.2)	1.5 (0.9–2.5)	21%**
2-OH-phenanthrene	2-PHEN	1.4 (0.8–2.2)	1.0 (0.6–1.4)	29%***	1.3 (0.8–2.2)	1.1 (0.5–1.3)	15%*	1.4 (0.7–2.2)	1 (0.6–1.4)	29%**
1-OH-pyrene	1-PYR	3.2 (2–3.9)	2.5 (1.6–3.6)	22%*	3.2 (2–3.7)	2 (1.5–3.4)	38%#	3.1 (2–5.2)	2.8 (1.7–4.2)	10%
Air measurements[a]		All, n = 44			Stove1, n = 26			Stove2, n = 18		
PM2.5-kitchen air (μg/m^3)		181 (78–402)	77 (39–129)	57%***	176 (83–390)	85 (49–129)	52%***	184 (68–414)	71 (17–136)	61%***
PM2.5-personal air (μg/m^3)		133 (69–191)	70 (47–97)	47%***	109 (62–190)	70 (48–97)	36%***	144 (89–215)	71 (29–105)	51%**
CO-kitchen air (ppm)		3.5 (1.2–5.9)	0.9 (0.3–1.9)	74%***	4.3 (1.4–6.7)	1.1 (0.4–1.9)	74%***	2.9 (1.2–5.3)	0.7 (0.3–1.8)	76%**
CO-personal air (ppm)		1.2 (0.5–2.1)	0.6 (0.2–1.2)	50%**	1.3 (0.7–2.3)	0.5 (0.1–1)	62%***	1.1 (0.3–1.9)	0.9 (0.2–1.3)	18%

#, $p<0.1$; *, $p<0.05$; **, $p<0.01$; ***, $p<0.001$, according to Wilcoxon matched pairs test.

[a] Fitzgerald (2009).

pyrene. We also measured $PM_{2.5}$ and CO levels in kitchen area air and personal air from the participants before and after the intervention as described elsewhere (Fitzgerald, 2009).

In the current investigation, the concentration of all ten urinary OH-PAH was significantly reduced in morning urine samples collected from the 57 participants three weeks after the installation of the new stoves. The largest median reduction in urine concentration after stove intervention was seen in 1 and 2-NAP, at 52% and 48%, respectively, while the metabolites of fluorene, phenanthrene and pyrene decreased by 19–33% (Table 1, Supplemental Material Figure S1). Urinary 1-PYR, the most commonly used biomarker for PAH exposure (Hansen et al., 2008), was 3.2 and 2.5 μg/g creatinine in pre and post-intervention, respectively, corresponding to a 22% reduction. The observed reduction in urinary level of 1- and 2-NAP was similar to that of $PM_{2.5}$ and CO in personal air (47% and 50% for $PM_{2.5}$ and CO, respectively). Reports have stated that exposure to naphthalene, a potential human carcinogen that exists almost exclusively in the gas phase in the ambient air, occurs primarily through inhalation (ATSDR, 2005; Preuss et al., 2003). Moreover, some theories have held that naphthalene in ambient air is associated primarily with indoor sources, such as cooking, heating and the use of naphthalene-containing moth repellents (ATSDR, 2005; Sanderson and Farant, 2004). Inhalation exposure of indoor air is the main exposure route to naphthalene in an non-occupationally exposed non-smoking population in the United States (Li et al., 2010). Therefore, the fact that the largest reduction in urinary levels was observed for metabolites from naphthalene is not surprising.

A limited number of studies used urinary biomarkers to evaluate indoor air pollution associated with biomass-burning or to evaluate the effectiveness of stove intervention programs. Torres-Dosal et al. (2008) reported urinary 1-PYR level in 20 residents from 10 households before and after a three-stage stove intervention program (removing indoor soot, paving dirt floors, and installing new stoves with chimney) in Mexico. Urinary 1-PYR was reduced from 13.0 μg/g creatinine to 9.3 μg/g creatinine, a 28.6% in reduction (Torres-Dosal et al., 2008). In a group of 412 Polish children aged 7–8 years, urinary 1-PYR levels were 1.09 μg/g creatinine in children living in households with coal-burning stove as domestic heating and cooking, which was significantly higher than the levels in those using non-biomass burning stoves (electric and gas, 0.78 μg/g creatinine), with a difference of 28.5% (Siwinska et al., 1999). In this study, we found that urinary 1-PYR levels were reduced by 22% after the stove intervention, similar to findings reported in the stove studies discussed above.

Total combined urinary PAH metabolite concentrations were correlated with $PM_{2.5}$ and CO in air during the pre-intervention phase (Supplemental Material Table S1), indicating indoor wood smoke as the common source for PAH, $PM_{2.5}$, and CO exposure, and inhalation as a major PAH exposure route in these subjects. Post-intervention testing also found positive correlations between urinary PAH biomarkers and air pollutants, and, as expected, the correlations were not as strong as in post-intervention phase, indicating higher relative contribution from other sources such as soot in the home and on cooking pots.

In our study, two separate stove programs were evaluated in order to study how and who to carry out the stove construction, knowledge which might be helpful designing and implementing future large-scale stove intervention projects. In Program A (Fig. 2B), subjects built their own stoves using materials received from the Juntos National Program, whereas in Program B (Fig. 2C), a contractor built all the stoves. No significant difference in levels of air pollutants and urinary PAH biomarkers was detected between these two stove programs ($p>0.1$), in the pre and post-intervention phases (Table 1). This finding is consistent with the fact that these two programs were carried out in communities with similar population, environmental settings, and lifestyles, and the performance of the intervention stoves were similar. Overall, Program A appeared to have a larger exposure reduction than Program B. Reduction in levels of all air pollutants and urinary biomarkers (with the exception of 1-PHEN) was significant or marginally significant in Program A, whereas in Program B, levels of personal CO and urinary 4-PHEN and 1-PYR were not significantly different between pre- and post-intervention (Table 1). This apparent difference may be a result of the limited sample size for both Programs. In addition, our study team could not access the Program B community because of rough terrain; therefore, field activities such as sample collection and oversight of new stove installation were conducted by trained local technicians. These factors could also affect the accuracy of the assessment in Program B.

It is encouraging to demonstrate significant reduction in human exposure to harmful wood smoke constituents by implementing improved stoves with relatively simple design. However, it should be noted that the urinary PAH levels in this study, even after the intervention, far exceeded the general population level in industrial countries as exemplified by a comparison to the National Health and Nutrition Survey 2001–2002 conducted in the United States (Fig. 3). Median concentrations of the fluorene, phenanthrene and pyrene metabolites post-intervention were higher than the 95th percentile in the U.S. population, and the naphthols were close to the 90th percentile (Fig. 3) (Li et al., 2008). Specifically, post-intervention median urinary 1-PYR level was 10-fold higher than the 95th percentile in the US population. Cigarette smoke is one of the major source for PAH exposure, especially in non-occupationally exposed population (Hecht, 2002; Scherer et al., 2000). Median level of urinary 1-PYR in the post-intervention phase is over 20-fold higher than that of smokers in the U.S. population (Li et al., 2008), over 8-fold higher than heavy smokers smoking over 20 cigarettes a day (Hecht et al., 2004), and higher than most other smoker levels reviewed by Hecht (2002), cf. Table 2. Those who work such jobs as coke oven worker, chimney sweeper, and aluminum worker are known to have a high exposure to PAHs and increased risk for developing lung, skin, and bladder cancer (IARC, 1984, 1985). Concentration of 1-PYR in this study is higher than that of asphalt pavers (Vaananen et al., 2003), comparable to or lower than that of coke oven workers (Simioli et al., 2004) and graphite electrode plant workers (Angerer et al., 1997), cf. Table 2. A cause for concern is that 16% of the subjects in this study had a post-intervention 1-PYR above 4.4 μg/g creatinine (2.3 μmol/mol creatinine), the proposed occupational exposure limit in the coke oven industry (Jongeneelen, 2001), and prior to intervention 23% had levels of 1-PYR above this exposure limit.

The high PAH exposure post intervention is most likely caused by partial release of wood smoke inside the houses, even with the use of the new stoves, and exposure to soot in the home or on cooking pots. We later discovered that local residents used long wood sticks/branches readily available from the mountains as their main fuel. The sticks could be fed into the stoves only partially, as shown in Fig. 2B, and a large amount of soot formed at the opening in front of the stove. This practice obviously reduced the efficiency of the intervention stove and illustrates the importance to understand local customs and practices before implementation of larger stove intervention programs. Further, biomass stoves do not necessarily improve the combustion efficiency and therefore cannot reduce the formation of harmful combustion products such as PAHs, $PM_{2.5}$ and CO. The chimney helped to lead large amount of wood smoke outdoor and thus reduced human exposure levels. However, the pollutants generated still contribute to ambient air pollution making biomass stove replacement programs more appropriate in rural areas.

New generations of stoves using advanced technology (e.g. battery powered fans) have been developed, boost burning efficiency and reducing hazardous combustion products (Mukunda et al., 2010), but those stoves are more expensive, making them cost-prohibitive in certain areas of the world, though they may be the only efficient choice of intervention in urban areas. Hence, the overall exposure situation as well as socioeconomic factors must be considered to design the most efficient stove intervention program possible for a certain area.

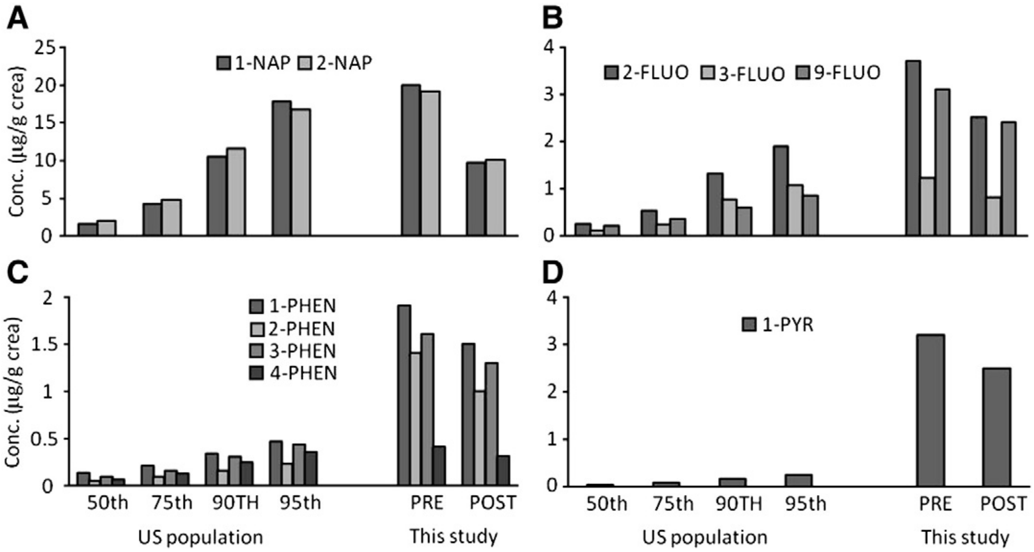

Fig. 3. Comparison of median OH-PAH concentrations pre and post the Peruvian stove intervention in this study to selected percentiles (50th, 75th, 90th, and 95th) found in the U.S. population.

Table 2
Comparison of median 1-hydroxypyrene (1-PYR) concentrations (creatinine-adjusted, µg/g creatinine) in selected studies.

Population	N	Median concentration (range)	Reference
Female adults, pre-stove intervention, Peru	57	3.2 (0.58–42)	This study
Female adults, post-stove intervention, Peru	57	2.5 (0.22–12)	This study
Children and adults, pre-stove intervention, Mexico	20	13 (2.1–34)[a]	Torres-Dosal et al. (2008)
Children and adults, post-stove intervention, Mexico	20	9.3 (0.7–35)[a]	Torres-Dosal et al. (2008)
Children with coal stove, Poland	194	1.09[b]	Siwinska et al. (1999)
Children with electric/gas stoves, Poland	218	0.78[b]	Siwinska et al. (1999)
US population	1625	0.041 (0.015–0.233)[c]	Li et al. (2008)
Smoker, US population	n/a	0.104[c]	Li et al. (2008)
Smokers with >20 cigarette/day	99	0.29[a]	Hecht et al. (2004)
Non-smoking coke oven workers, Italy	31	4.5 (0.48–46)	Simioli et al. (2004)
Graphite electrode plant workers, Germany	67	8.7 (0.2–326)	Angerer et al. (1997)
Non-smoking asphalt pavers, Finland	26	0.46 (0.16–4.2)[a]	Vaananen et al. (2003)

[a] Geometric mean.
[b] Least square geometric mean.
[c] Median concentration with 10th and 95th percentiles.

5. Conclusions

The improved stoves with chimney significantly reduced human exposure to hazardous combustion products including PAHs, $PM_{2.5}$ and CO. However, even after the intervention, urinary OH-PAH levels in these subjects were still far exceeding that of general population in the United States, higher than smokers, and at comparable levels to workers with known high occupational exposure to PAHs. . Multiple factors, ranging from costs to local and cultural customs, must be considered in future stove improvement programs.

Disclaimer

The co-authors of this manuscript do not have any financial conflict of interest with any parties that were involved in this study. The findings and conclusions in this report are those of the authors and do not necessarily represent the Centers for Disease Control and Prevention.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.envint.2011.03.024.

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