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PM_{2.5} in household kitchens of Bhaktapur, Nepal, using four different cooking fuels



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HIGHLIGHTS

- One of the largest databases of indoor PM2.5 measurements from cookstoves.
- PM_{2.5} levels were measured in kitchens using low-cost nephelometers.
- The nephelometers results were well correlated with results from gravimetric method.
- Decreasing PM_{2.5} was associated with biomass, kerosene and then LPG/electric stoves.
- PM_{2.5} levels in the kitchens with electric stoves were similar to ambient PM_{2.5} levels.

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ABSTRACT

In studies examining the health effects of household air pollution (HAP), lack of affordable monitoring devices often precludes collection of actual air pollution data, forcing use of exposure indicators, such as type of cooking fuel used. Among the most important pollutants is fine particulate matter ($PM_{2.5}$), perhaps the best single indicator of risk from smoke exposure. In this study, we deployed an affordable and robust device to monitor PM_{2.5} in 824 households in Bhaktapur, Nepal. Four primary cooking fuels were used in roughly equal proportions in these households: electricity (22%), liquefied petroleum gas (LPG) (29%), kerosene (23%), and biomass (26%). PM_{2.5} concentrations were measured in the kitchens using a light-scattering nephelometer, the UCB-PATS (University of California, Berkeley-Particle and Temperature monitoring System). The major predictors of PM_{2.5} concentrations in study households were investigated. The UCB-PATS results were well correlated with the gravimetric results ($R^2 = 0.84$; for all fuels combined). The mean household PM_{2.5} concentrations across all seasons of the year were 656 (standard deviation (SD):924) μg/m³ from biomass; 169 (SD: 207) μg/m³ from kerosene; 101 (SD: 130) $\mu g/m^3$ from LPG; and 80 (SD: 103) $\mu g/m^3$ from electric stoves. In the multivariate regression of PM_{2.5} measures, compared with electric stoves, use of LPG, kerosene and biomass stoves were associated with increased indoor PM_{2.5} concentrations of 65% (95% CI: 38-95%), 146% (103-200%), and 733% (589 -907%), respectively. The UCB-PATS performed well in the field. Biomass fuel stoves without flues were the most significant sources of PM_{2.5}, followed by kerosene and then LPG stoves. Outdoor PM_{2.5}, and season influenced indoor PM_{2.5} levels. Results support careful use of inexpensive light-scattering monitors for monitoring of HAP in developing countries.

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Abbreviations: ALRI, acute lower respiratory infection; CI, confidence interval; HAP, household air pollution; LPG, liquefied petroleum gas; PCs, particle coefficients; PM $_{2.5}$, particles less than 2.5 μ m in diameter; PM $_{10}$, particles less than 10 μ m in diameter; UCB-PATS, University of California, Berkeley-Particle and Temperature monitoring System; WHO, World Health Organization.

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1. Introduction

Globally, about 2.8 billion people use unprocessed solid fuels (coal or biomass) for cooking in poor populations mostly in developing countries (Bonjour et al., 2013). LPG and kerosene are also commonly used in these same areas as household fuels (Mills, 2005). These cooking fuels are usually burned in unvented stoves and in poorly ventilated kitchens. Several epidemiology studies have suggested that exposure to solid fuel and kerosene smoke increases disease risk (Bates et al., 2013; Lim et al., 2012; Pokhrel et al., 2010). The vast majority of epidemiological studies investigating the association between cooking fuels and health effects in developing countries, however, have used surrogates of household air pollution (HAP) exposure, such as type of fuel used, kitchen location or ventilation (Dherani et al., 2008). The difficulty and expense associated with a large number of air pollution measurements in household settings have been the main reasons to use exposure proxies in these studies. Among the most important, but difficult, pollutants to measure is fine particulate matter (PM_{2.5}), thought to be the single best indicator of health risk from combustion smoke (Naeher et al., 2007). Various devices are available to measure indoor PM2.5 levels but they are either expensive or will not measure up to PM levels found in high pollution settings, as occur in developing-country households (Northcross et al., 2010).

Progress has been made to develop less-expensive devices to measure PM_{2.5} in household settings. In particular, the UCB-PATS (University of California, Berkeley-Particle and Temperature monitoring System) has been used in small HAP monitoring studies around the world (Alnes et al., 2014; Armendariz-Arnez et al., 2010; Chengappa et al., 2007; Chowdhury et al., 2007b; Clark et al., 2011; Gurley et al., 2013a; Masera et al., 2007; Northcross et al., 2010; Sanbata et al., 2014). See web supplement.

Here the UCB-PATS was used to measure indoor $PM_{2.5}$ concentrations in households from a case—control study examining associations between HAP and acute lower respiratory infection (ALRI) in children ≤ 3 years, resident in Bhaktapur municipality, in the Kathmandu Valley, Nepal. Health effects by stove/fuel type are described elsewhere (Bates et al., 2013).

Using the UCB-PATS adjusted for local conditions, the main objectives of the study reported here were: 1) to determine concentrations of PM_{2.5} in households using four different cooking fuels; 2) to identify demographic and household predictors of PM_{2.5} concentration; and 3) to determine the influence of outdoor PM_{2.5} concentrations on indoor concentrations.

2. Material and methods

2.1. Human subjects

Human Subjects' approvals were obtained from institutional review boards at the University of California, Berkeley, and at the Institute of Medicine, Tribhuvan University Teaching Hospital, Kathmandu, Nepal. Field work was conducted May 2006 to June 2007.

2.2. Study site

The study was conducted in Bhaktapur Municipality, about 13 km east of Kathmandu, the capital of Nepal. Bhaktapur is a combined rural/urban area with approximately 70,000 residents (Bates et al., 2013).

The most common cooking fuels in Bhaktapur are kerosene, LPG, wood, rice-husks and electricity. Kerosene is used either in

wick or pressure stoves. Biomass is used in a traditional open fire mud stove, called a *Chulo*, with 1–3 potholes, sometimes with a chimney. Rice-husk metal stoves without a chimney or hood are also commonly used. Many households had an electric stove with an open coil on a mud frame/base.

Bhaktapur has a sub-tropical, temperate climate, and four distinct seasons: winter (December to February), pre-monsoon/spring (March to May), monsoon/summer (June to September) and post-monsoon/autumn (October to November) (Giri et al., 2008). During the winter, the temperature sometimes drops to 0 °C. During this season, space heating with biomass, LPG, coal or electricity is common.

Contributing outdoor air pollution, there is one major highway and about 10–12 brick kilns on the outskirts of the municipality. There are many narrow brick roads in the interior of the municipality, mainly traveled by motorcycles and tractors.

2.3. Study participants and household selection for PM_{2.5} measurements

 $PM_{2.5}$ levels were measured in households of participants of the case—control study previously described (Bates et al., 2013). These participants were recruited from an open cohort of children ≤ 3 years old, under active surveillance for respiratory illness (Mathisen et al., 2009, 2010). Eligible participants were recruited from Siddhi Memorial Hospital, where children were brought for ALRI consultation. After ALRI confirmation, fieldworkers took consent from the child's caregiver and scheduled the HAP measurements. Simultaneously, controls, age-matched by month, were recruited from a list of children under surveillance, but without ALRI at the time. After informed consent, HAP was measured in control houses.

All HAP measurements were done within a week of recruitment. Altogether, 917 children (452 cases and 465 controls) were recruited, of which HAP was measured in 824 households (393 cases and 431 controls). HAP measurements began one month after the first recruitment of cases and controls. Therefore, we did not measure $PM_{2.5}$ in the first 40 homes (24 cases and 16 controls), nor later in another 53 homes (35 cases and 18 controls) due to malfunctioning of the air pollution monitors.

2.4. HAP measurements

HAP measurement involved two steps. First, UCB-PATS were gravimetrically calibrated in a subset of 54 households to derive local particle coefficients (PCs) for processing future measurements. PCs for wood, rice husk and LPG fuels were derived by plotting UCB-PATS mass (in millivolts) against the gravimetric mass ($\mu g/m^3$) in least-squares regression (PC: β_0 , the intercept parameter of the regression line). For kerosene and electric stoves, we used the mean values (\sum (gravimetric mass/UCB-PATS Delta mVolts)/n) to derive PCs.

Gravimetric $PM_{2.5}$ levels were also measured in some children's bedrooms (n = 9), and monitors were placed on the rooftops (~15 m above ground) of eight children's homes to measure outdoor $PM_{2.5}$. These homes were on the same elevation (~1400 m above sea level), and were at least 2–3 km away from the highway and brick kilns, and within 1 km of the government air quality monitoring station. No tall trees or higher structures around these houses obstructed the airflow. Gravimetric $PM_{2.5}$ samples were collected over 24 h using air sampling pumps (Model 224-PCXR8, SKC Inc., PA) with $PM_{2.5}$ sharp-cut cyclones (scc1.062 BGI Triplex Cyclone, BGI Inc., Waltham, MA) on a 37-mm diameter Teflon filter (2 μ m pore size, PTFE Ring) at a flow rate of 1.5 l/min. All pumps were calibrated using a rotameter prior to and after each sampling

period. All filters were weighed (pre- and post-sampling) by the same person (AP) at the University of California, Berkeley. Filters were weighed on a six-digit Mettler Toledo MT-5 balance (SN 1118413759). Filters were conditioned for at least 24 h in a temperature- and humidity-controlled room before weighing. The temperature was maintained at 17-23 °C, and relative humidity kept below 42%. During the weighing, polonium²¹⁰ alpha source stripes were used to eliminate interference of electrostatic charges on the filters. Lab blank filters were weighed before every weighing session to compare the weight measured during the session to the historically available measurements. Filters were always weighed three times, and average values used to calculate the mass. Blank filters were both left in the lab and taken to the field and were weighed to correct the filter mass. The average change in weight of field blanks was (0.000005 mg). As this was negligible, no blank subtraction was done.

In children's bedrooms, pumps were placed at sleeping height. Results from the co-located monitors were used to estimate separate field-based particle coefficients for wood, rice husks, kerosene, LPG and electric stoves.

UCB-PATS were placed in study houses in the same locations as during the validation—at 1.5 m height and about 1 m from the edge of the main stove and at least 1 m from any doors or other openings in the walls. Data reported here are based on 24 h of monitoring (within one minute) on weekdays, in which the start and end time of monitoring was ~9:00 AM on successive days. Each UCB-PATS was zeroed by placing it inside a particle-free plastic (Ziploc) bag for 30—60 min before and after each monitoring period. Particle and temperature coefficients and the results from zeroing were subsequently used in the data processing. The light-scattering sensing chamber of each UCB-PATS was cleaned every two weeks with a wipe and 70% isopropanol. UCB-PATS batteries were replaced when the voltage dropped below 7.5 V.

2.5. Questionnaires

HAP measurements were accompanied by administration of pre- and post-monitoring questionnaires. These questionnaires collected three groups of variables: 1) variables for the day of monitoring, such as type of primary and secondary stoves used in the house, unusual stove use pattern during the HAP monitoring period, weather conditions during the monitoring period, ventilation in the kitchen (e.g., open doors and windows) and other smoke exposure sources, such as number of smokers in the house, use of incense or mosquito coils; 2) fixed variables, such as kitchen size or the presence of roads within 100 m; and 3) variables describing usual practices, such as types of non-electric lamp used when power is unavailable, and type of space heating used during the winter. Participants' caregivers/parents were also asked about their occupations and household characteristics, such as construction materials.

2.6. Outdoor PM₁₀ and meteorological data

The Nepal Government operates a central air pollution monitoring station in Bhaktapur, to measure ambient PM_{10} . Measurements for the study period (May, 2006 to May, 2007) were obtained from the Ministry of Environment (251 days of measurement). Using gravimetric $PM_{2.5}$ data measured on rooftops of the eight households of the participants in the UCB-PATS validation study, we calculated the outdoor $PM_{2.5}$ to PM_{10} ratio for May, 2006, when the validation study was conducted. Outdoor $PM_{2.5}$ measurements were conducted during a non-rain period. Then, matching by days of indoor and outdoor PM measurements, we estimated ambient $PM_{2.5}$ levels for the entire study period by multiplying measured outdoor

 PM_{10} by the $PM_{2.5}/PM_{10}$ ratio. We also evaluated the influence of outdoor $PM_{2.5}$ levels on indoor $PM_{2.5}$ levels by subtracting estimated outdoor $PM_{2.5}$ levels from indoor/kitchen $PM_{2.5}$ levels.

2.7. Statistical analysis

First, arithmetic mean concentrations, standard deviations (SD) and 95% confidence intervals (95% CI) were calculated. PM2 5 concentrations were examined according to primary and secondary fuel-stove types, season, demographic factors, energy use-related behaviors and household characteristics, using analysis of variance (ANOVA). Then PM_{2.5} data were observed graphically and logtransformed for modeling. To identify covariates that influenced the PM_{2.5} concentration, normal error bivariate regression models were run between log (linear)-transformed PM_{2.5} concentration data and particular covariates. Any covariates associated with PM_{2.5} concentrations at $p \le 0.20$ were considered as potential predictors for use in the multivariate regression model. This p-value represents a balance between specificity and sensitivity and is likely to capture any important confounders (Maldonado and Greenland, 1993). In addition, we used a simple directed acyclic graph (DAG) approach to check whether any covariates lay on the causal pathway between the main exposure of interest-primary fuel stove type-and the outcome-PM_{2.5} concentration. Any covariates on the causal pathway were not included as predictors in the multivariate model (Greenland et al., 1999).

In the ANOVA, and regression analyses, the wood and rice husk stove categories were combined as the 'biomass stove' category.

The normal error multivariate linear regression model was used to assess sources and significant predictors of PM_{2.5} concentration in participants' houses. The regression model was run on the log (linear)-transformed PM_{2.5} concentration data with the Huber/ White/Sandwich linearized estimator (robust standard error option). This option does not assume that variance is constant and controls the chance of violation of assumption of constant variance and normality of residuals in the multivariate regression. This gives the most realistic estimates of the variance and the robust standard errors for the parameter estimates (Cameron and Trivedi, 2009). After running the model, multi-collinearity among regression parameters was checked using a post-regression command, variation inflation factor (VIF) (StataCorp, 2007). Conventionally, a VIF greater than 10 indicates multi-collinearity (Cameron and Trivedi, 2009). To confirm that residuals of regression models were normally scattered around zero, residuals versus fitted values were plotted in Q-norm plots. Results are reported in non-transformed (arithmetic) format, because these are more easily interpretable in terms of health risks.

Model specification errors can substantially affect the estimate of regression coefficients. To check this, the post-regression Ramsey regression specification error test (Ramsey RESET) was conducted (Hamilton, 2006). The Ramsey RESET tests whether the model includes all relevant variables and excludes irrelevant variables. Small p-values (<0.05) indicate model specification errors (omitted variables could be significant predictors). The results obtained from the multivariate regression model were normalized by exponentiating the regression coefficients (exp $^\beta$). Percent change in PM_{2.5} exposure was estimated with the algorithm: ([exp $^\beta-1$]x 100) (Baumgartner et al., 2011; Introduction to SAS, 2014). STATA version 12 (StataCorp LP, TX, USA) was used for all analyses.

3. Results

3.1. PM_{2.5} concentration by primary and secondary fuel stove type

Table 1 shows the distribution of households by primary and

secondary stove types and kitchen average $PM_{2.5}$ concentrations ($\mu g/m^3$). Study participants had roughly equal proportions of the four primary stove types—biomass (26%) [wood, 21%, rice-husks, 5%], kerosene (23%), LPG (29%) and electric (22%). About 43% of study participants reported use of a secondary stove. No-one reported using a rice-husk stove as a secondary stove.

The kitchen PM_{2.5} concentrations had a log-normal distribution for all stove types. The 24-h arithmetic mean PM_{2.5} concentrations associated with rice-husk stoves (759 $\mu g/m^3$) and wood stoves (630 $\mu g/m^3$) [wood & rice-husk combined-biomass 656 $\mu g/m^3$] were highest, and lower for electric stoves (80 $\mu g/m^3$), LPG (101 $\mu g/m^3$) and kerosene stoves (169 $\mu g/m^3$) (ANOVA p < 0.05).

We evaluated the joint distribution of average kitchen PM_{2.5} concentrations by primary and secondary stove types across households (Table 2). Primary biomass stove users were more likely to have an electric stove as a secondary stove (39%) than were primary kerosene stove users (7%).

Average kitchen $PM_{2.5}$ concentrations were highest in households where both primary and secondary fuel stove types used biomass (811 μ g/m³). Concentrations were also relatively high when only the secondary stove used biomass (Table 2).

3.2. PM_{2.5} concentration by household characteristics

The differences in $PM_{2.5}$ concentrations by demographic, behavioral and household characteristics are presented in Table 3. Kitchen levels of $PM_{2.5}$ varied significantly by both parents' occupations and reported daily stove use in hours (p < 0.05). Having an open door or window while cooking (ventilation) with a biomass stove was associated with decreased $PM_{2.5}$ concentrations (p < 0.05). Use of indoor lighting devices (kerosene lamps or candles) during power outages, and smokers living in the house (p < 0.05) were associated with increased $PM_{2.5}$ concentrations. Other variables, such as unusual stove use during monitoring, use of incense or mosquito coils, types of wall and roof in the kitchen, and motorable road within 100 m, showed an opposite trend.

3.3. Seasonal effect on kitchen $PM_{2.5}$ levels

During the study period, mean indoor temperatures recorded by the UCB-PATS were 14 $^{\circ}$ C in December (winter) and 26 $^{\circ}$ C in June (summer). The mean ambient temperatures recorded at the government meteorological station were 4 $^{\circ}$ C in December/January and 30 $^{\circ}$ C in June. The minimum and maximum daily rainfalls during the study period were 0 and 44 mm and the daily mean was

4 mm.

In all seasons, $PM_{2.5}$ concentrations were highest in association with biomass fuel stoves and lowest with either electric or LPG stoves. Table 4 summarizes the results of $PM_{2.5}$ concentration by fuel/stove type and season.

3.4. Outdoor gravimetric PM_{10} and predicted $PM_{2.5}$, and kitchen $PM_{2.5}$ concentrations before and after the subtraction of outdoor $PM_{2.5}$

Minimum and maximum outdoor PM_{10} levels measured at the central air pollution monitoring station were 11 and 139 $\mu g/m^3$, with an annual average of 68 (SD: 33) $\mu g/m^3$.

The average gravimetric $PM_{2.5}$ concentration measured outside the 8 homes, in May 2006, was 45 (SD:26) $\mu g/m^3$. The average gravimetric outdoor PM_{10} concentration in the same month was 82 (SD:22) $\mu g/m^3$, giving an ambient $PM_{2.5}$ to PM_{10} ratio of 0.55.

Annual ambient PM_{10} concentrations in Bhaktapur showed a seasonal cyclic pattern with higher concentrations during winter $(94 \,\mu\text{g/m}^3)$ and pre-monsoon $(92 \,\mu\text{g/m}^3)$ seasons, and lower during the monsoon $(46 \,\mu\text{g/m}^3)$ and autumn $(77 \,\mu\text{g/m}^3)$.

Table 5 shows the average kitchen $PM_{2.5}$ concentration before and after the subtraction of estimated outdoor $PM_{2.5}$. It also provides ratios of indoor to outdoor $PM_{2.5}$ in kitchens by the main stove types.

Predicted minimum and maximum outdoor PM_{2.5} levels were 6 and 76 µg/m³, with an annual average of 37 (SD:18) µg/m³. Seasonal average values were 51 (SD:15) µg/m³ in the pre-monsoon; 25 (SD:15) µg/m³ in the monsoon; 42 (SD:9) µg/m³ in the autumn; and 52 (SD:10) µg/m³ in the winter period. The mean kitchen PM_{2.5} concentrations associated with biomass, kerosene, LPG and electric stoves after the subtraction of estimated outdoor PM_{2.5} levels were 649 (SD:1051) µg/m³, 135 (SD:214) µg/m³, 55 (SD:113) µg/m³ and 31 (SD:96) µg/m³, respectively.

The arithmetic mean indoor (kitchen)/outdoor (I/O) mass concentrations of $PM_{2.5}$ before subtraction of estimated outdoor $PM_{2.5}$ were greater in the biomass stove-using kitchens (I/O ratio: 24) followed by kerosene (ratio: 7), and LPG stove-using kitchens (ratio: 4). The mean I/O ratio in the kitchens using electric stoves was about 2.

3.5. Bivariate and multivariate regression

In bivariate linear regression models, based on measured PM_{2.5} concentrations without subtracting estimated ambient

Table 1 Distribution of average kitchen $PM_{2.5}$ concentration in $\mu g/m^3$ (SD) across households by primary and secondary stove type for cooking.

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Stove fuel type	N (%) of households	Arithmetic mean kitchen PM _{2.5} concentration in µg/m³ (SD)	95% CI	Geometric mean kitchen PM _{2.5} concentration in µg/m ³	95% CI
Primary fuel stove type					
Wood	174 (21)	630 (908)*	495-766	356	305-415
Rice husk	44 (5)	759 (988)	459-1060	505	389-655
Biomass (Wood + Rice husk)	218 (26)	656 (924)*	533-780	382	334-437
Kerosene	187 (23)	169 (207)	139-199	117	105-131
LPG	238 (29)	101 (130)	84-117	72	66-79
Electric stove	181 (22)	80 (103)	65-95	55	49-62
Secondary fuel stove type					
No secondary stove	468 (57)	218 (431)	178-257	110	100-120
Wood	95 (12)	219 (348)	148-290	98	77-125
Kerosene	94 (11)	270 (710)	125-416	106	84-135
LPG	32 (4)	131 (215)	53-208	73	51-103
Electric stove	135 (16)	451 (838)	308-593	209	172-255

ANOVA * p value < 0.05.

Note: ANOVA test comparing the mean PM_{2.5} concentrations across primary and secondary fuel stove types.

Table 2 Joint distribution of arithmetic mean kitchen PM_{2.5} concentrations by primary and secondary stove types across households, Bhaktapur, Nepal.

Secondary fuel stove type	Primary fuel stove type								
	Mean PM _{2.5} concentration in $\mu g/m^3$ (SD) and frequency (n)								
	Biomass	Biomass Kerosene LPG					Electric		
	N (%)	μg/m³(SD)	N (%)	$\mu g/m^3$ (SD)	N (%)	μg/m³ (SD)	N (%)	μg/m³ (SD)	
Biomass	11 (5)	811 (564)	11 (6)	324 (398)	6(3)	176 (283)	67 (37)	109 (146)	
Kerosene	25 (11)	782 (1250)	2(1)	60 (32)	25 (11)	97 (63)	42 (23)	79 (89)	
LPG	8 (4)	346 (355)	1(1)	253 (.)	5(2)	61 (33)	18 (10)	48 (25)	
Electric	85 (39)	648 (1003)	13 (7)	173 (170)	37 (16)	95 (73)		_	
No secondary stove	89 (41)	638 (810)	160 (86)	158 (190)	165 (69)	101 (141)	54 (30)	56 (36)	

concentrations (Table 6), $PM_{2.5}$ concentrations in the kitchen varied significantly ($p \le 0.20$) by fathers' and mothers' occupation, duration of stove use in years, use of a secondary stove, ventilation in the kitchen, use of an alternative lighting source in electricity outages, use of space heating, number of smokers, type of roof and wall in the kitchen, and season.

In the multivariate model (Table 6) when all covariates associated with the $PM_{2.5}$ concentrations at $p \le 0.20$ in bivariate linear regression models were included, compared with primary electric stove use, use of LPG, kerosene, and biomass primary stoves were associated with increased $PM_{2.5}$ concentrations by 65% (95% CI: 38, 95%), 146% (95% CI:103, 200%), and 733% (95% CI:589, 907%), respectively. Use of biomass-burning secondary stoves was associated with an increase in the $PM_{2.5}$ concentration by 43% (95% CI:14, 80%). Compared with the pre-monsoon/spring, the $PM_{2.5}$ concentrations in the winter increased by 45% (95% CI:20, 75%). Mother's occupation as housewife was associated with a 20% increase in $PM_{2.5}$ (95% CI:4, 38%). Overall, the model explained 50% of variation in $PM_{2.5}$ concentration.

The average variance inflation factor (VIF) for the final model was 1.77 (range 1.07, 3.56). As a VIF greater than 10 suggests multicollinearity of the independent variables, collinearity between regressors in the final model should not be of concern. Similarly, the model specification error test (Ramsey RESET test) had a p-value of 0.97, suggesting that specification error in the model was not of concern. The Q-norm plots showed the residuals of the regression close to a normal distribution. Table 6 lists percent change in PM_{2.5} based on log regression coefficient estimates for the multivariate regression of indoor PM_{2.5} concentrations.

4. Discussion

This analysis involves one of the largest databases of indoor $PM_{2.5}$ measurements from cookstoves with a balanced distribution of fuel types.

The UCB-PATS and gravimetric correlations determined by the coefficient of determination R² in the validation study, and duplicate measurements are in the range of previously reported correlations for indoor environments using other instruments (Chowdhury et al., 2007a; Edwards et al., 2006). The UCB-PATS and gravimetric correlations were generally better than reported correlations between commercial light-scattering devices (nephelometers) and gravimetric instruments, and somewhat comparable to reported correlations between optical particle counters (OPC) and gravimetric methods used in other studies (Giorio et al., 2013; Quintana et al., 2000; Weber et al., 2012; Wu et al., 2004). This provides confidence that the UCB-PATS is a useful tool for largescale monitoring of household air pollution in developing countries. However, it is important to conduct co-location experiments in local conditions to generate suitable mass calibration factors for the aerosol mixtures of interest. See web supplement.

This study provides evidence that both indoor and outdoor sources contribute to indoor PM_{2.5}, but indoor sources predominate in kitchens that use biomass or kerosene stoves. Outdoor PM2.5 sources are much more influential in kitchens that use LPG or electric stoves (see Table 5). Unsurprisingly, kitchen PM_{2.5} levels were highest for biomass stove homes and lowest for electric stove homes. PM_{2.5} levels in electric stove homes were relatively close to the ambient PM_{2.5} measurements, although nearby outdoor measurements were not taken every day and so an exact relationship could not be defined. Predicted outdoor PM_{2.5} concentrations for Bhaktapur were generally consistent with levels measured in Kathmandu, 13 km west of Bhaktapur (Aryal et al., 2009). For example, outdoor PM_{2.5} levels during 2006-2007 in Kathmandu have been reported as 69 μ g/m³ in the pre-monsoon, 30 μ g/m³ in the monsoon, 53 $\mu g/m^3$ in the post-monsoon season and 90 $\mu g/m^3$ in the winter (Arval et al., 2009). The predicted outdoor PM_{2.5} levels for Bhaktapur during these time periods were 51 (SD:15) μg/m³ in the pre-monsoon; 25 (SD:15) μ g/m³ in the monsoon; 42 (SD:9) μ g/ m^3 in the post-monsoon; and 52 (SD:10) $\mu g/m^3$ in the winter.

In the multivariate regression model, only primary and secondary stove types, mother's occupation and winter season, appeared as significant ($p \leq 0.05$) predictors of PM_{2.5} concentration. Ventilation factors–opening of windows during cooking and only an open door during cooking–showed some association with decreased PM_{2.5} concentration. Most of these associations were not statistically significant.

The mean $PM_{2.5}$ concentration documented from biomass stoves in this study is similar to the results of other indoor air pollution monitoring studies in Nepal. For example, in a study conducted near the Kathmandu Valley, using photometric devices similar to the UCB-PATS, one-day mean indoor concentrations of $PM_{2.5}$ of $792 \mu g/m^3$ (range $136-2610 \mu g/m^3$) from biomass fuel stoves were reported (Kurmi et al., 2008).

We also found a strong seasonal effect on indoor PM2.5 concentration, especially higher concentrations during the winter. This finding is consistent with other studies conducted in Asia (Baumgartner et al., 2011; Gurley et al., 2013b). The higher concentrations of PM_{2.5} during the winter may be attributable to the burning of more biomass (for heating) and/or reduced ventilation in the household. UCB-PATS data loggers recorded a mean indoor temperature of 14 °C in December and January, with a minimum of 0 °C on some days. Generally, windows and doors are kept closed in winter, but in the warmer monsoon and summer seasons, they are kept mostly open. Another possible explanation for the strong seasonal effect on indoor PM_{2.5} concentrations, especially during the winter, could be the contribution of ambient sources to indoor air. Ambient PM concentrations in the Kathmandu valley, are highest in winter, possibly in part because brick kilns operate then (Aryal et al., 2009).

We found kitchen PM_{2.5} levels were higher in households where mothers reported house work as their main occupation–possibly

 Table 3

 Comparison of arithmetic mean kitchen PM_{2.5} concentration in $\mu g/m^3$ (SD) by demographics, stove type, and behavioral and household characteristics.

Characteristics	N (%)	All	Biomass	Kerosene	LPG	Electric stove
Variables determined on the day of	monitoring					
Father's occupation						
Self-employed & salary earner	308 (37)	185 (340)*	540 (625)	138 (95)*	106 (146)	74 (107)
Factory/daily wage worker	416 (51)	268 (533)	646 (870)	165 (209)	84 (78)	84 (107)
Other	100 (12)	448 (943)	838 (1309)	362 (402)	121 (149)	81 (78)
Mother's occupation						
Outside home	621 (75)	235 (455)*	623 (747)	143 (126)*	91 (97)*	73 (101)*
House work	203 (25)	331 (762)	754 (1312)	207 (287)	147 (224)	112 (110)
Age of stove (years)						
≤1	63 (8)	273 (611)	1090 (1224)	173 (251)	130 (170)	54 (26)
>1 and ≤3	151 (18)	210 (563)	875 (1387)	189 (222)	76 (68)	92 (154)
≥3 and ≤5	67 (8)	218 (406)	656 (689)	101 (53)	127 (229)	58 (25)
>5	543 (66)	275 (551)	608 (857)	170 (209)	103 (120)	83 (99)
Daily stove use in hours						
≤2	573 (70)	246 (493)*	613 (823)	164 (193)	96 (131)	87 (111)
2 to ≤3	181 (22)	345 (750)	809 (1187)	198 (268)	116 (143)	72 (101)
>3	70 (8)	139 (248)	449 (553)	123 (93)	101 (88)	49 (35)
Kitchen ventilation						
Both door and window/s open	686 (83)	255 (451)	604 (693)*	167 (202)	103 (138)	80 (94)
Only door open	65 (8)	320 (872)	824 (1420)	140 (78)	96 (74)	110 (211)
Only window/s open	66 (8)	259 (987)	4236 (5295)	189 (263)	92 (111)	35 (17)
Neither door nor window/s open	7 (1)	56 (16)	_	_	64 (19)	47 (6)
Number of smokers in the house						
0	324 (39)	199(454)*	651 (940)	161 (231)	104 (154)	71 (93)
1	402 (49)	266 495)	615 (778)	178 (208)	95 (89)	83 (95)
>2	98 (12)	424 (898)	770 (1221)	148 (91)	106 (105)	95 (161)
Type of factory inside the house						
None	681 (83)	240 (437)**	587 (703)*	174 (223)	103 (130)	80 (104)***
Carpet & mill	100 (12)	323 (857)	858 (1434)	122 (65)	98 (154)	52 (44)
Other	43 (5)	396 (1010)	1283 (1887)	177 (143)	81 (52)	136 (171)
Exhaust in the kitchen						
Chimney	13 (1.6)	137 (110)	173 (138)	207 (90)	116 (.)	29 (7)
Exhaust fan	2 (0.2)	465 (455)	787 (.)	144 (.)	_ ``	_ ` `
None	809(98.2)	260 (552)	663 (931)	168 (210)	101 (130)	81 (104)
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Characteristics	N (%)	All (n = 824) Mean (SD)	Biomass	Kerosene	LPG	Electric stov
Unusual stove use during monitoring		200 (572)	(550 (650)	450 (404)*	405 (400)	TO (00)**
None	709 (86)	260 (572)	679 (979)	159 (191)*	105 (138)	73 (90)**
Cooked for more people than usual	76 (9)	227 (307)	484 (416)	126 (103)	80 (50)	135 (182)
Cooked for less people than usual	29 (4)	298 (461)	645 (752)	344 (354)	53 (24)	110 (98)
Other	10 (1)	268 (455)	675 (859)	290 (443)	58 (12)	31 (20)
Weather during monitoring	101 (00)		=== (,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	.=. (0.=)	100 (100)	== (==)
No rain	491 (60)	284 (652)	753 (1110)***	171 (217)	103 (136)	73 (85)
Light rain	151 (18)	222 (369)	559 (576)	128 (80)	86 (115)	83 (108)
Rain	182 (22)	218 (309)	467 (446)	187 (237)	105 (124)	99 (147)
Fixed variables						
Wall in the kitchen						
No wall	7 (0.8)	462 (442)***	587 (470)	_	_	153 (133)
Porous ^a & other wall	12 (1.5)	537 (664)	699 (700)	76 (.)	_	41 (6)
Solid wall	805 (97.7)	253 (546)	656 (943)	169 (21)	101 (130)	79 (104)
Roof in the kitchen						
Concrete	357 (43.3)	176 (397)*	552 (986)	165 (217)	101 (128)	72 (94)
Metal sheet	178 (21.6)	348 (637)	620 (822)	166 (107)	99 (152)	67 (57)
Wood and mud	285 (34.6)	304 (634)	750 (1004)	179 (190)	100 (116)	87 (119)
Others	4 (0.5)	379 (360)	491 (344)	_	_	43 (.)
Kitchen size						
Large or medium Small	557 (67.6)	256 (468)	612 (738)	174 (229)	102 (131)	81 (102)
or very small	266 (32.3)	265 (686)	766 (1272)	160 (163)	96 (126)	78 (106)
Missing	1 (0.1)	66 (.)	_	_	66 (.)	_
Motorable road within 100 m						
None	58 (7)	271 (596)	912 (1160)	184 (231)	75 (43)	73 (34)***
Highway	93 (11)	254 (386)	501 (562)	222 (297)	119 (184)	40 (23)
Main road	162 (20)	244 (389)	678 (614)	160 (155)	84 (92)	121 (176)
Small road	511 (62)	263 (608)	658 (1020)	150 (186)	105 (132)	75 (93)
	, ,		, ,		• • • • • • • • • • • • • • • • • • • •	· , ,
Characteristics N ((n = 824) an (SD)	Biomass	Kerosene	LPG	Electric stov
Variables determined about usual p						
Light when electricity fails	4 (4) 14	5 (323)*	867 (906)	153 (56)	74 (43)	56 (43)***
Light when electricity fails Battery lamp or none 34		, ,	, ,	, ,	, ,	, ,
Light when electricity fails Battery lamp or none 3- Candle 45	9 (56) 17	5 (323)* 3 (287) 3 (780)	867 (906) 506 (489) 737 (1090)	153 (56) 164 (229) 175 (182)	74 (43) 107 (142) 78 (49)	56 (43)*** 69 (71) 98 (136)

Table 3 (continued)

Characteristics	N (%)	All (n = 824) Mean (SD)	Biomass	Kerosene	LPG	Electric stove
Incense or mosquito coi	l use					
No	507 (62)	290 (689)***	719 (1157)	168 (204)	115 (163)***	70 (88)
Yes	317 (38)	239 (437)	612 (713)	169 (210)	92 (105)	86 (112)
Space heating stove who	en it is cold					
None	680 (82.5)	257 (562)	678 (967)	166 (202)	92 (111)**	75 (90)***
Electric stove	20 (2.4)	76 (67)	310 (.)	_	72 (44)	52 (33)
Kerosene	2 (0.2)	240 (261)	424 (.)	_	56 (.)	_
LPG	4 (0.3)	130 (167)	_	_	162 (188)	34 (.)
Firewood	118 (14.3)	306 (517)	582 (749)	187 (255)	154 (209)	123 (174)

ANOVA * p value < 0.05; **p value < 0.10 ***p value < 0.20.

Note: ANOVA test comparing the mean PM_{2.5} concentrations across demographics, primary stove fuel type, behavioral and household characteristics.

Table 4 Comparison of average kitchen $PM_{2.5}$ concentration ($\mu g/m^3$) by main stove type and season.

Stove/Fuel	Spring	Pre-monsoon/Summer	Autumn	Winter	ANOVA p-value
Biomass					
n	28	131	35	24	0.59
Arithmetic mean (95% CI)	675 (249-1100)	601 (489-713)	698 (334-1060)	877 (206-1550)	
SD	1100	649	1060	1588	
Range	76-5750	63-4220	78-6300	67-7980	
Geometric mean (95% CI) Kerosene	372 (189–369)	362 (304–432)	426 (309–687)	445 (282–702)	
n	39	101	20	27	0.21
Arithmetic mean (95% CI)	153 (121-185)	165 (121-209)	120 (76-165)	240 (124-356)	
SD	100	224	96	292	
Range	45-472	31-1440	41-385	52-1100	
Geometric mean (95% CI) LPG	128 (106–155)	110 (94–128)	96 (71–130)	152 (108–215)	
n	51	118	34	35	0.01
Arithmetic mean (95% CI)	85 (61-108)	94 (72-116)	78 (53-104)	167 (93-241)	
SD	83	120	74	216	
Range	19-593	21-75	25-456	24-1010	
Geometric mean (95% CI)	69 (58-81)	66 (58-75)	64 (53-78)	114 (87-149)	
Electric stove					
n	44	76	22	39	0.49
Arithmetic mean (95% CI)	77 (4-110)	69 (44-94)	95 (51-140)	96 (69-124)	
SD	111	109	100	84	
Range	12-548	15-775	18-501	22-383	
Geometric mean (95% CI)	48(37-63)	48 (41-57)	69 (49-98)	73 (58-92)	

Table 5 Comparison of average kitchen (indoor) and outdoor $PM_{2.5}$ concentration ($\mu g/m^3$) by main stove type.

Stove/Fuel	Kitchen PM _{2.5} concentration	Predicted outdoor PM _{2.5} concentration	Kitchen PM _{2.5} — Outdoor PM _{2.5}	Ratio (kitchen PM _{2.5} /Outdoor PM _{2.5})
Biomass				
n	218	170	154 ^a	154
Arithmetic mean (95% CI)	656 (533-780)	36 (33-39)	649 (482-816)	24.32 (18.62-30.02)
SD	924	18	1051	35.79
Geometric mean (95% CI)	382 (334-437)	31 (29-34)	316 (261-382)	12.19 (10.20-14.57)
Kerosene				
n	187	163	137 ^b	137
Arithmetic mean (95% CI)	169 (139-198)	37 (34-40)	135 (99-172)	6.47 (4.58-8.35)
SD	207	18	214	11.15
Geometric mean (95% CI)	117 (105-131)	32 (29-35)	73 (59–88)	3.69 (3.15-4.31)
LPG				
n	238	181	163 ^c	163
Arithmetic mean (95% CI)	101 (84-117)	38 (35-40)	55 (37-72)	3.56 (2.64-4.47)
SD	130	18	113	5.89
Geometric mean (95% CI)	72 (66–79)	33 (30-36)	36 (29-45)	2.09 (1.82-2.40)
Electric stove				
n	181	137	127 ^d	127
Arithmetic mean (95% CI)	80 (65-95)	39 (36-42)	31 (14-48)	2.33 (1.80-2.86)
SD	103	19	96	3.03
Geometric mean (95% CI)	55 (49-62)	33 (30-37)	25 (19-34)	1.49 (1.26-1.75)

^a 16 outdoor or kitchen PM_{2.5} concentration data not available.

^a Several openings on wall.

 ¹⁶ outdoor or kitchen PW_{2.5} concentration data not available.
 26 outdoor or kitchen PM_{2.5} concentration data not available.
 18 outdoor or kitchen PM_{2.5} concentration data not available.
 10 outdoor or kitchen PM_{2.5} concentration data not available.

Table 6Coefficient estimates for bivariate and multivariate linear regression of independent variables on indoor concentrations of PM_{2.5} without subtracting predicted outdoor PM_{2.5} concentration

Characteristics	Bivariate regression analysis	Multivariate regression analysis		
	% Change in PM2.5 (95% CI) based on log regression	% Change in PM2.5 (95% CI) based on log regression		
Primary stove				
Electric stove	Reference	Reference		
LPG	31 (13,51)	65 (38, 95)		
Kerosene	112 (82,148)	146 (103, 200)		
Biomass	589 (481, 725)	733 (589, 907)		
Duration of stove use in years				
<1	Reference	Reference		
1-3	-11(-36, 25)	-12 (-31, 12)		
3-5	-4(-34,40)	-14(-35, 14)		
>5	22 (-10, 67)	-9 (-27, 15)		
Secondary stove				
No secondary stove	Reference	Reference		
Electric stove	92 (54, 136)	-5 (-21, 14)		
LPG	-34(-53, -6)	-16(-36, 9)		
Kerosene	-3(-24,25)	11 (-9, 34)		
Biomass	-10 (-31, 15)	43 (14, 80)		
Ventilation in the kitchen		. , ,		
Both door and window/s open	Reference	Reference		
Only door open	7 (-21, 43)	7 (-14, 34)		
Only window/s open	-24(-41,-1)	2 (-18, 27)		
Neither door nor window/s open	-55 (-64,-44)	-17 (-37, 11)		
Number of smokers in the house	,			
0	Reference	Reference		
1	36 (16, 58)	6(-7,20)		
>2	88 (43, 146)	9 (-11, 35)		
Father's occupation				
Self-employed & salary earner	Reference	Reference		
Factory/daily wage worker	31 (12, 54)	1 (-10, 13)		
Others	75 (32, 132)	13 (-9, 40)		
Mother's occupation				
Outside home	Reference	Reference		
House work	36 (15, 62)	20 (4, 38)		
Wall in the kitchen		, , ,		
No wall	Reference	Reference		
Porous and others	-10 (-72, 183)	3 (-61, 169)		
Solid wall	-56 (-81, 3)	-2(-54, 112)		
Roof in the kitchen	, ,	,		
Concrete	Reference	Reference		
Metal sheet	46 (19, 79)	-10 (-24, 7)		
Wood & mud	27 (7, 51)	14 (-1, 32)		
Others	127 (-27, 603)	1 (-56, 127)		
Season	• • •			
Pre-monsoon-spring	Reference	Reference		
Monsoon-summer	22 (1, 49)	-2 (-16, 13)		
Autumn	31 (1,70)	6 (-12, 28)		
Winter	42 (11,80)	45 (20, 75)		

related to more cooking indoors.

In the multivariate regression, ventilation and construction materials did not appear as significant predictors of $PM_{2.5}$ concentrations, although coefficients were usually in expected directions. Having a metal sheet roof in kitchens showed some association with decreased $PM_{2.5}$ concentration; and a wood or mud roof in the kitchen had a non-statistically significant association with increased $PM_{2.5}$ concentrations. Compared with wood or mud roofs, thatched or metal sheet roofs permit ventilation in the kitchen through eaves. Lower PM_{10} concentrations have been reported in Bangladesh kitchens with thatched or corrugated iron roofs, compared with mud roofs (Dasgupta et al., 2006).

Another indicator of ventilation–opening doors or windows while cooking did not appear as a significant predictor of kitchen PM_{2.5} concentrations. This is consistent with some other studies (Menon, 1988). Other studies, however, have shown an effect of ventilation in reducing indoor PM concentration (Baumgartner et al., 2011; Bruce et al., 2004; Dasgupta et al., 2006; Gurley et al., 2013b). For example, in rural China never opening windows or

doors was associated with 75% (95% CI:0%—201%) higher PM concentration relative to always ventilating the kitchen (Baumgartner et al., 2011). We asked only about the usual practice of opening windows or doors while cooking in the kitchen. We did not ask detailed questions about frequency of opening over the entire day or during cooking.

This study provides evidence that biomass stoves, followed by kerosene and then LPG stoves are most strongly associated with PM_{2.5} concentrations in Bhaktapur kitchens. Biomass-burning secondary stoves also contribute substantial PM_{2.5}.

The multivariate regression model explained 50% of the variation of indoor $PM_{2.5}$ concentrations. At least some of the remaining variation is likely to be accounted for imperfect specification of variables, including cooking time, other household combustion sources, and ventilation factors during the measurement periods.

Our study had some limitations. First, in the validation study, there were only three kerosene and two electric stoves. To avoid the possibility of random error from too few data points, we did not determine R² between the UCB-PATS and the gravimetric method

for fuel types for which there were fewer than five data points. Instead we used the mean values to derive PCs to correct the UCB mass. In the case of kerosene stoves, however, there was not much difference in PCs determined by the mean and the R² method (0.015 vs. 0.016 mean values, respectively). The corresponding mean values for electric stoves were 0.007 and 0.012. Thus, if we had used the PCs based on the R² method then we would have estimated slightly less PM_{2.5} mass for the kerosene and the electric stoves.

A second limitation was that we could not measure outdoor gravimetric PM_{2.5} in all seasons; instead we estimated annual ambient PM_{2.5} levels using results of gravimetric PM_{2.5} measurements conducted during the summer/pre-monsoon season. PM₁₀ and PM_{2.5} can be highly spatially variable and have seasonal differences, for which we had no data for Bhaktapur. Also, it is likely that there will have been some filtering of the air as it passed inside. Thus, the assumptions of a constant PM_{2.5}/PM₁₀ ratio across seasons and complete indoor infiltration will almost certainly have been sources of exposure misclassification in Table 5. However, our main analysis (Table 6) used results unadjusted for estimated ambient PM_{2.5} results.

Another limitation was that reporting of some of the household characteristics, such as kitchen size, unusual stove use, and rain pattern during the monitoring, were subjective. Although answers to these questions were based on observations by the study staff, we cannot rule out the possibility of exposure misclassification. It is difficult to assess the direction and impact of such misclassifications.

5. Conclusion

Compared with Nepal's national 24-h indoor air quality standard for $PM_{2.5}$ (60 $\mu g/m^3$), without taking into account ambient concentrations, the mean PM_{2.5} concentrations in kitchens that used biomass, kerosene, LPG and electricity were, respectively, 11, 2.8, 1.7 and 1.3 times higher. Even after subtracting the estimated ambient contribution, the mean PM_{2.5} concentrations in kitchens that used these fuels were, respectively, 11, 2.3, 0.9 and 0.5 times the national 24-h indoor air quality standard (Ministry of Science Technology and Environment (2009)). All the averages and nearly every house measured exceeded the WHO Air Quality Guideline Interim-Target I (WHO IT-1) annual level of 35 μg/m³ PM_{2.5} (World Health Organization, 2005). If, however, we assume that the estimated outdoor PM_{2.5} concentrations reflect the ambient contribution to kitchen concentrations, after subtracting those concentrations, households using electric stoves (average 31 µg/ m³) would be in compliance with the Nepal standard and most households would meet the WHO IT-1 level. LPG is considered to be a clean cooking fuel (Global Alliance for Clean Cookstoves, 2014). However, even after subtracting estimated ambient PM_{2.5} concentrations, PM_{2.5} concentrations in kitchens using LPG were higher than the WHO IT-1 concentration. At these relatively low levels, however, it is difficult to distinguish the particles from the food being cooked from those due to combustion processes. Therefore, this study's results supports a recommendation that a hood be used to minimize the cook's exposure from use of any cookstove.

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The views expressed herein are those of the authors and do not necessarily reflect the views of the institutions where they work.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.04.060.

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