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Chemical Composition of $PM_{2.5}$ in Wood Fire and LPG Cookstove Homes of Nepali Brick Workers

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Abstract: Household air pollution is a major cause of morbidity and mortality worldwide, largely due to particles ≤ 2.5 µm ($PM_{2.5}$). The toxicity of $PM_{2.5}$, however, depends on its physical properties and chemical composition. In this cross-sectional study, we compared the chemical composition of $PM_{2.5}$ in brick workers' homes (n = 16) based on use of wood cooking fire or liquefied petroleum gas (LPG) cookstoves. We collected samples using RTI International particulate matter (PM) exposure monitors (MicroPEMs). We analyzed filters for 33 elements using energy-dispersive X-ray fluorescence and, for black (BC) and brown carbon (BrC), integrating sphere optical transmittance. Wood fire homes had significantly higher concentrations of BC (349 µg/m^3) than LPG homes (6.27 µg/m^3, $p < 0.0001$) or outdoor air (5.36 µg/m^3, $p = 0.002$). Indoor chlorine in wood fire homes averaged 5.86 µg/m^3, which was approximately 34 times the average level in LPG homes (0.17 µg/m^3, $p = 0.0006$). Similarly, potassium in wood fire homes (4.17 µg/m^3) was approximately four times the level in LPG homes (0.98 µg/m^3, $p = 0.001$). In all locations, we found aluminum, calcium, copper, iron, silicon, and titanium in concentrations exceeding those shown to cause respiratory effects in other studies. Our findings suggest the need for multi-faceted interventions to improve air quality for brick workers in Nepal.

Keywords: household air pollution; fine particulate matter; international environmental health; cookstove; respiratory disease; brick worker

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1. Introduction

Household air pollution from the indoor burning of solid fuels, such as wood, crop residues, dung, or coal, is associated with 3.8 million deaths annually worldwide [1,2]. Exposure to household air pollution is associated with low birth weight, asthma, chronic obstructive pulmonary disease (COPD), respiratory infections, impaired immune function, coronary heart disease (CHD), stroke, cataracts, and cancers, including lung cancer [3–6]. Among household air pollutants generated from solid fuels, particulate matter (PM) less than or equal to 2.5 microns (µm) in aerodynamic diameter ($PM_{2.5}$), also called fine particulate matter, may be the single largest contributor to this excess disease burden [7,8]. However, the toxicity of $PM_{2.5}$ appears to be partially dependent on its chemical composition, which varies widely based on local emission sources [9–16].

In the Kathmandu Valley, Nepal, there are over 30,000 seasonal brick workers [17]. Most brick workers in Nepal live on-site at the brick kiln [18]. The most common type of housing for these workers is brick huts with tin roofs, often with poor ventilation to the outdoors [19]. Within this population of workers, the two primary methods of cooking are with indoor open wood fires or with liquefied petroleum gas (LPG) cookstoves [18,19].

Previous studies by our group found that Nepali brick workers, in addition to having hazardous work-related respiratory exposures [20], experience significant $PM_{2.5}$ exposures during non-working hours [18,19]. Indoor $PM_{2.5}$ concentrations in brick workers' homes with wood fire and LPG cookstoves were 541.14 and 79.32 $\mu g/m^3$, respectively, and these elevated levels coincided with meal and sleep times [19]. Brick workers suffer a disproportionate burden of respiratory symptoms compared to other workers in the same community [21], and we propose that these symptoms may be partially explained by elevated $PM_{2.5}$ levels in brick workers' homes, particularly among those cooking indoors with open wood fires.

$PM_{2.5}$ generated during wood combustion is composed primarily of elemental (EC) and organic carbon (OC), nitrate and sulfate species, metals, and other elements [22]. However, wood smoke composition depends on the species of wood being burned and the burn temperature [22–24]. Low burn temperatures (300–500 °C), such as during the start-up phase of an open wood fire, generally produce larger particles composed of numerous OC species and low levels of trace elements and metals, while higher burn temperatures (>800 °C), such as during the burn phase of an open wood fire, produce smaller particles composed of higher EC/OC ratios, and higher levels of trace elements and metals [8,23].

The biological mechanisms behind many of the diseases associated with wood smoke inhalation are not well understood, but studies suggest some metals, metalloids, and nonmetal elements may play key roles in air pollution-related diseases [9]. For example, studies of $PM_{2.5}$ constituents in ambient air pollution reported that the elements aluminum (Al), calcium (Ca), chlorine (Cl), iron (Fe), nickel (Ni), titanium (Ti), vanadium (V), and zinc (Zn), as well as black carbon (BC), are associated with increased hospitalizations and mortality, particularly among people $\geq$65 years of age [10,11]. The metals copper (Cu), Fe, potassium (K), and Zn, and the metalloid silicon (Si), are associated with respiratory hospital admissions in children, with the most serious effects seen in those $\leq$5 years of age [12]. The metals Al, Ni, Zn, V, and Ti, and the metalloid Si, are associated with low birth weight [13,14]. Several of these elements are present in wood smoke, in varying concentrations, depending on the species of wood and the burn temperature [22,24–26]. BC is associated with increased morbidity and mortality, primarily from heart and lung diseases [15]. Brown carbon (BrC), another constituent of $PM_{2.5}$ found in areas where the use of solid biomass fuels is high [27], may also influence human health because it can attach to toxic chemicals, such as benzopyrene, and heavy metals [28].

Understanding the chemical composition of $PM_{2.5}$ among specific populations may help elucidate relationships between exposure and disease. Our previous study measured the chemical composition of $PM_{2.5}$ in brick workers' homes during daytime hours when most home occupants were working, and thus did not capture pollutants generated during non-working hour activities such as cooking [18]. The purpose of this study, therefore, was to measure the chemical composition of $PM_{2.5}$ over a full day in order to characterize non-working hour exposures.

2. Materials and Methods

2.1. Study Design

We collected $PM_{2.5}$ samples using both filter-based and real-time nephelometer methods. Our previous study reported the $PM_{2.5}$ total mass and nephelometer trend analyses [19]. For this study, we analyzed the 25 mm 3.0 µm PTFE filters (Zefon International, Ocala, FL, USA) for 35 chemical constituents. The methods for home selection, measures of housing characteristics, and air filter handling and sampling strategy, described briefly here, are described in full in our previously published paper [19]. We used a cross-sectional study design to measure $PM_{2.5}$ constituents in brick workers' homes (n = 17) from a single brick kiln in Bhaktapur, Nepal. We recruited homes by convenience sampling, and we classified them as either wood cooking fire or LPG cookstove homes. The typical construction of the homes sampled in this study was detailed previously [19]. We collected samples from 30 April to 3 May 2019 for approximately 21 h (median: 21.21; interquartile range: 2.21) in each home. We administered an extant questionnaire [18], by means of an interpreter, to measure housing factors, including number of people in the home, number of children in the home, primary fuel used for cooking, presence of smokers in the home, and the number of smokers in the home. We also measured the living area of the home and calculated the occupant density as the number of occupants divided by the home area in m^2. Prior to data collection, Brigham Young University's (BYU) Institutional Review Board (IRB) determined that this study did not meet the definition of human subject research, per 45 CFR 46 [29], based on the fact the unit of study was the home rather than the individual.

2.2. Indoor and Outdoor $PM_{2.5}$ Measurements

We collected $PM_{2.5}$ samples using MicroPEM V.3.2A personal exposure monitors (RTI International, Research Triangle Park, NC, USA), which we placed indoors, on a tripod, approximately 1.2 m from the floor. Simultaneous daily outdoor samples were collected on-site at the brick kiln in a centralized location. Detailed methods describing MicroPEM preparation, placement, and filter handling were described previously [19].

2.3. Elemental Analysis

RTI International performed the analysis of the 25 mm filters for 33 elements following the IO3.3 compendium method [30], which was modified for use with the Thermo (Thermo Fisher Scientific, Waltham, MA, USA) ARL energy-dispersive X-ray fluorescence instrument equipped with a silicon drift detector. This instrument configuration was used because it could produce enough spectral counts to fully quantify each element, while collimating the beam. The instrument was calibrated with thin-film standards (Micromatter Technologies Inc., Surrey, BC, Canada) that approximated PM deposition on a filter and the unknown samples were analyzed under identical excitation conditions. The samples were analyzed under vacuum with five different energy conditions to achieve maximum sensitivity, while avoiding overlapping spectra. A camera system within the instrument chamber was used to ensure the beam was focused on the exposed area of the filter to accurately quantify the elements of concern. A multi-element thin film standard was analyzed with each tray of samples to ensure there was acceptable instrument performance across the mass range and to assess instrument drift.

2.4. Carbon Analysis

Following gravimetric analysis, all sample filters were shipped to RTI International for optical analysis using RTI International's integrating sphere optical transmittance technique [31]. The optical transmittance through the filter and the deposited PM sample were measured at seven wavelengths, ranging from near-infrared (940 nm) to blue (430 nm). All the sample filter transmittance data were adjusted using the mean transmittance of 10 blank filters from the same manufacturer's lot. An empirically-derived algorithm used the measured wavelength-dependent transmittance values to quantify the BC and lightly absorbing BrC contributions to the total PM collected on the sample filter. This technique, and similar optical methods, have been used in numerous PM exposure studies as a low-cost and non-destructive means of obtaining basic PM compositional data from sample filters [32–36].

2.5. Statistical Analyses

All statistical analyses were conducted using SAS version 9.4 (SAS Institute, Inc., Cary, NC, USA). Although we collected 20 total $PM_{2.5}$ samples, the filter of one sample tore and could not be analyzed for $PM_{2.5}$ chemical components. Thus, we excluded that sample from all statistical analyses. We used $\alpha = 0.05$ as the significance level for all analyses.

We calculated the frequencies and percentages for categorical characteristics of homes at the brick kiln and arithmetic means, standard deviations, minimums, first quartiles, medians, third quartiles, and maximums for the continuous characteristics of the homes at the brick kiln. For $PM_{2.5}$ chemical components, we calculated the frequency and percentage of samples that had concentrations below the lower detection limits (LDL), at or between the LDLs and upper detection limits (UDL), and above the UDLs. We also calculated the geometric means (GM), 95% confidence intervals (CI), minimums, and maximums for concentrations of $PM_{2.5}$ chemical components. We used GMs because the distributions of the concentrations of almost all $PM_{2.5}$ chemical components were right-skewed. For individual $PM_{2.5}$ chemical components that had all concentrations at or between the LDLs and UDLs, we used separate intercept-only linear regression models, with the natural logarithm of concentrations of individual $PM_{2.5}$ chemical components as the dependent variables, and then exponentiated intercept coefficients to calculate GMs and 95% CIs. For individual $PM_{2.5}$ chemical components with some concentrations below the LDLs or above the UDLs, we used separate intercept-only Tobit regression models, with the natural logarithm of concentrations of individual $PM_{2.5}$ chemical components as the dependent variables, and then exponentiated intercept coefficients to calculate GMs and 95% CIs.

We used decision rules that were similar to those of Beard at al. [37], who based their decision rules on information from Lubin et al. [38], to determine the appropriate types of regression models to use for the analyses of individual $PM_{2.5}$ chemical components with varying proportions of concentrations at or between the LDLs and UDLs. For the individual $PM_{2.5}$ chemical components that had all concentrations at or between the LDLs and UDLs, we estimated the *p*-values and unadjusted associations between the individual characteristics of homes at the brick kiln and the concentrations of individual $PM_{2.5}$ chemical components, using separate simple linear regression models, with the natural logarithm of the concentrations of individual $PM_{2.5}$ chemical components as the dependent variables. For the individual $PM_{2.5}$ chemical components that had >30–99% of concentrations at or between the LDLs and UDLs, we estimated the *p*-values and unadjusted associations between the individual characteristics of homes at the brick kiln and the concentrations of individual $PM_{2.5}$ chemical components, using separate simple Tobit regression models, with the natural logarithm of the concentrations of individual $PM_{2.5}$ chemical components as the dependent variables. For individual $PM_{2.5}$ chemical components that had >0–30% of concentrations at or between the LDLs and UDLs, we estimated the *p*-values and unadjusted associations between individual characteristics of homes at the brick kiln and the concentrations of individual $PM_{2.5}$ chemical components, using separate simple exact unconditional logistic regression models, with dichotomous indicator variables (i.e., one if the concentration was $\geq$LDL and zero if the concentration was <LDL) as dependent variables. For each of the three types of regression models, we exponentiated slope coefficients to calculate GMs, geometric mean ratios (GMR), or exact odds ratios (OR) and 95% CIs.

We considered several versions (e.g., linear; linear and quadratic; linear, quadratic, and cubic; natural logarithm; and categorical) of continuous characteristics of homes at the brick kilns and used the versions that had the lowest values of the Akaike Information Criterion (AIC) [39,40]. Where appropriate, we conducted pairwise comparisons of the GMs of concentrations of $PM_{2.5}$ chemical components for each category of home area and fuel type and location and used the Tukey (linear regression models) or Tukey-Kramer (Tobit regression models) method to adjust the *p*-values for multiple comparisons. We estimated multivariable linear or Tobit regression models when more than one characteristic of homes at the brick kilns were statistically significantly associated with concentrations of a particular $PM_{2.5}$ chemical component. For sensitivity analyses, we repeated analyses using home volume instead of home area.

3. Results

3.1. Characteristics of Homes of Brick Workers

For the 16 homes that we collected $PM_{2.5}$ samples from (i.e., excluding the one home for which the filter tore), the home area was 5.41–9.50 m^2 for 38%, >9.50–10.67 m^2 for 31%, and >10.67–31.40 m^2 for 31% (Table 1). The mean number of people in the home was 3.31 and the mean occupant density was 33.54 people per 100 m^2. Sixty-three percent of homes had 0–1 children and 38% had 2–3 children. Fifty percent of homes had smokers and the median number of smokers in the home was 0.5. Sixty-nine percent of homes used LPG for fuel and 31% used wood.

Table 1. Characteristics of homes [a] at a brick kiln in Bhaktapur, Nepal (May 2019).

Characteristic	Homes, n (%)	Mean	SD	Min	Q1	Median	Q3	Max
Total	16 (100)							
Home area [b], m^2								
5.41–9.50	6 (38)							
>9.50–10.67	5 (31)							
>10.67–31.40	5 (31)							
Number of people in home		3.31	1.54	1.00	2.00	3.00	4.00	7.00
Occupant density, number of people/100 m^2		33.54	17.00	9.55	22.47	30.11	38.55	73.96
Number of children in home								
0–1	10 (63)							
2–3	6 (38)							
Smokers in home								
No	8 (50)							
Yes	8 (50)							
Number of smokers in home		0.75	1.06	0.00	0.00	0.50	1.00	4.00
Fuel type								
LPG	11 (69)							
Wood	5 (31)							

Abbreviations: LPG, liquefied petroleum gas; Max, maximum; Min, minimum; Q1, first quartile; Q3, third quartile; and SD, standard deviation. [a] The filter of one sample tore and could not be analyzed, and, thus, was excluded from analyses. [b] Categories based on tertiles.

3.2. Summary Statistics for $PM_{2.5}$ Chemical Component Concentrations

Six $PM_{2.5}$ chemical components had all concentrations below the LDLs (Table 2). For the other 29 $PM_{2.5}$ chemical components, GMs of the concentrations ranged from 0.000042 μg/m^3 for $PM_{2.5}$ cerium to 16.09 μg/m^3 for $PM_{2.5}$ BC, with a median GM of 0.016 μg/m^3 for $PM_{2.5}$ barium (Ba).

Table 2. Summary statistics for the mean of samples inside or outside homes [a] at a brick kiln in Bhaktapur, Nepal (May 2019).

					Between LDL and UDL					
$PM_{2.5}$ Chemical Component, μg/m^3	LDL Mass (μg)	LDL Concentration Range	Missing, n	Below LDL, n (%)	n (%)	GM [b]	95% CI [b]	Min [c]	Max [c]	Above UDL
Al	0.012	0.018, 0.052		2 (11)	17 (89)	0.30	0.14, 0.66	0.031	2.19	0 (0)
Sb	0.24	0.34, 1.01		19 (100)	0 (0)	NA	NA	NA	NA	0 (0)
As	0.0024	0.0034, 0.010		17 (89)	2 (11)	0.0031	0.0025, 0.0039	0.0039	0.0039	0 (0)
Ba	0.0047	0.0068, 0.020		7 (37)	12 (63)	0.016	0.0083, 0.032	0.010	0.10	0 (0)
BC	0.50	0.73, 2.15		1 (5)	14 (74)	16.09	5.82, 44.52	1.84	107.36	4 (21) [d]
Br	0.0021	0.0030, 0.0089		0 (0)	19 (100)	0.022 [e]	0.016, 0.030 [e]	0.0078	0.061	0 (0)
BrC	0.50	0.73, 2.15	4	0 (0)	15 (100)	10.56 [e]	7.89, 14.13 [e]	2.34	17.88	0 (0)
Cd	0.082	0.12, 0.35		19 (100)	0 (0)	NA	NA	NA	NA	0 (0)
Cs	0.0024	0.0034, 0.010		6 (32)	13 (68)	0.0077	0.0050, 0.012	0.0048	0.024	0 (0)
Ca	0.0022	0.0033, 0.0096		0 (0)	19 (100)	0.18 [e]	0.075, 0.42 [e]	0.0060	1.33	0 (0)
Ce	0.0024	0.0034, 0.010		18 (95)	1 (5)	0.000042	0.0000000052, 0.35	0.015	0.015	0 (0)
Cl	0.0019	0.0028, 0.0082		0 (0)	19 (100)	0.38 [e]	0.14, 1.07 [e]	0.023	17.13	0 (0)
Cr	0.0013	0.0019, 0.0055		10 (53)	9 (47)	0.0020	0.0013, 0.0032	0.0019	0.0086	0 (0)
Co	0.00096	0.0014, 0.0041		15 (79)	4 (21)	0.00093	0.00047, 0.0019	0.0015	0.0032	0 (0)
Cu	0.0016	0.0023, 0.0067		6 (32)	13 (68)	0.0043	0.0025, 0.0076	0.0025	0.048	0 (0)
In	0.12	0.18, 0.52		19 (100)	0 (0)	NA	NA	NA	NA	0 (0)
Fe	0.0016	0.0024, 0.0071		0 (0)	19 (100)	0.26 [e]	0.12, 0.57 [e]	0.022	1.69	0 (0)
Pb	0.0049	0.0071, 0.021		6 (32)	13 (68)	0.014	0.0088, 0.021	0.0099	0.11	0 (0)
Mg	0.0050	0.0072, 0.021		5 (26)	14 (74)	0.032	0.016, 0.065	0.011	0.20	0 (0)
Mn	0.0018	0.0025, 0.0075		6 (32)	13 (68)	0.0083	0.0040, 0.017	0.0034	0.054	0 (0)
Mo	0.012	0.017, 0.051		19 (100)	0 (0)	NA	NA	NA	NA	0 (0)
Ni	0.0010	0.0015, 0.0043		10 (53)	9 (47)	0.0017	0.0013, 0.0023	0.0020	0.0038	0 (0)

Table 2. *Cont.*

PM$_{2.5}$ Chemical Component, µg/m^3	LDL Mass (µg)	LDL Concentration Range	Missing, n	Below LDL, n (%)	Between LDL and UDL n (%)	GM [b]	95% CI [b]	Min [c]	Max [c]	Above UDL
P	0.0024	0.0036, 0.011		5 (26)	14 (74)	0.011	0.0061, 0.021	0.0045	0.064	0 (0)
K	0.0019	0.0028, 0.0083		0 (0)	19 (100)	1.44 [e]	0.94, 2.18 [e]	0.38	5.86	0 (0)
Rb	0.0023	0.0034, 0.010		9 (47)	10 (53)	0.0043	0.0022, 0.0081	0.0043	0.028	0 (0)
Se	0.0022	0.0033, 0.0097		12 (63)	7 (37)	0.0033	0.0026, 0.0041	0.0039	0.0070	0 (0)
Si	0.0064	0.0093, 0.027		0 (0)	19 (100)	1.08 [e]	0.58, 2.00 [e]	0.14	5.15	0 (0)
Ag	0.055	0.079, 0.24		19 (100)	0 (0)	NA	NA	NA	NA	0 (0)
Na	0.010	0.015, 0.045		2 (11)	17 (89)	0.13	0.088, 0.19	0.034	0.37	0 (0)
Sr	0.0030	0.0043, 0.013		17 (89)	2 (11)	0.0015	0.00025, 0.0088	0.0075	0.0092	0 (0)
S	0.0026	0.0038, 0.011		0 (0)	19 (100)	2.77 [e]	2.20, 3.48 [e]	0.99	4.79	0 (0)
Sn	0.18	0.26, 0.76		19 (100)	0 (0)	NA	NA	NA	NA	0 (0)
Ti	0.00085	0.0012, 0.0036		2 (11)	17 (89)	0.021	0.0088, 0.050	0.0018	0.19	0 (0)
V	0.0011	0.0015, 0.0046		9 (47)	10 (53)	0.0021	0.0012, 0.0039	0.0020	0.013	0 (0)
Zn	0.0015	0.0022, 0.0066		0 (0)	19 (100)	0.059 [e]	0.039, 0.089 [e]	0.019	0.39	0 (0)

Abbreviations: Al, aluminum; Sb, antimony; As, arsenic; Ba, barium; BC, black carbon; Br, bromine; BrC, brown carbon; Cd, cadmium; Cs, cesium; Ca, calcium; Ce, cerium; Cl, chlorine; Cr, chromium; Co, cobalt; CI, confidence interval; Cu, copper; GM, geometric mean; In, indium; Fe, iron; Pb, lead; LDL, lower detection limit; Mg, magnesium; Mn, manganese; Max, maximum; Min, minimum; Mo, molybdenum; Ni, nickel; NA, not applicable; PM$_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 µm; P, phosphorus; K, potassium; Rb, rubidium; Se, selenium; Si, silicon; Ag, silver; Na, sodium; Sr, strontium; S, sulfur; Sn, tin; Ti, titanium; UDL, upper detection limit; V, vanadium; and Zn, zinc. [a] The filter of one inside sample tore and could not be analyzed, and so that home was excluded from analyses. Of the remaining 19 samples, 16 were from inside and three were from outside the homes. [b] Estimated via simple (i.e., unadjusted), intercept only Tobit regression models of the natural logarithm transformed values. [c] Calculated from samples that had values at or between the LDL and UDL. [d] The UDL mass was 80 µg and the UDL concentration range was 116.08 to 344.21 µg/m^3. [e] Estimated via simple (i.e., unadjusted), intercept only linear regression models of the natural logarithm transformed values.

3.3. *Associations between Characteristics of Homes of Brick Workers and PM$_{2.5}$ Chemical Component Concentrations*

Home area was significantly associated with concentrations of PM$_{2.5}$ Cl (p = 0.03) and PM$_{2.5}$ Cu (p = 0.005; Supplementary Materials, Table S1). For PM$_{2.5}$ Cl, pairwise comparisons indicated significant differences between 5.41–9.50 m^2 (GM = 2.95 µg/m^3) and >10.67–31.40 m^2 (GM = 0.14 µg/m^3; p = 0.04), but not between 5.41–9.50 m^2 and >9.50–10.67 m^2 (GM = 0.25 µg/m^3; p = 0.10) or >9.50–10.67 m^2 and >10.67–31.40 m^2 (p = 0.85). For PM$_{2.5}$ Cu, pairwise comparisons indicated significant differences between 5.41–9.50 m^2 (GM = 0.013 µg/m^3) and >9.50–10.67 m^2 (GM = 0.0024 µg/m^3; p = 0.01), and between 5.41–9.50 m^2 and >10.67–31.40 m^2 (GM = 0.0035 µg/m^3; p = 0.03), but not between >9.50–10.67 m^2 and >10.67–31.40 m^2 (p = 0.82). The number of people in the home, occupant density, and the number of children in the home were not significantly associated with concentrations of any PM$_{2.5}$ chemical component (Supplementary Materials, Tables S1 and S2). The presence of smokers in the home was significantly associated with concentrations of 22 of 29 (76%) PM$_{2.5}$ chemical components, and the GMs of concentrations were higher in homes with smokers than in homes without smokers for all 22 significant associations (Table 3). Similarly, the number of smokers in the home was significantly associated with concentrations of 20 (69%) PM$_{2.5}$ chemical components and the GMRs were greater than one for all 20 significant associations (i.e., the GMs of the concentrations of those 20 PM$_{2.5}$ chemical components increased as the number of smokers in the home increased). Fuel type and location was significantly associated with concentrations of 22 (76%) PM$_{2.5}$ chemical components (Table 4). Pairwise comparisons indicated significant differences between LPG, indoor, and wood, indoor, for 21 (95%) of the significant associations, and the GMs of concentrations were higher for wood, indoor, than for LPG, indoor, for all 21 significant differences. Pairwise comparisons indicated significant differences between LPG, indoor, and outdoor for one (5%) of the significant associations (i.e., PM$_{2.5}$ BrC) and the GM of concentrations was higher for LPG, indoor, than for outdoor. Pairwise comparisons indicated a significant difference between wood, indoor, and outdoor for six (27%) of the significant associations, and the GMs of concentrations were higher for wood, indoor, than for outdoor for all six significant differences.

Table 3. Associations between the mean of samples inside homes [a] and smokers in the home and the number of smokers in the home at a brick kiln in Bhaktapur, Nepal (May 2019).

	Smokers in Home					Number of Smokers in Home		
	No		Yes					
$PM_{2.5}$ Chemical Component, μg/m^3	GM [b]	95% CI [b]	GM [b]	95% CI [b]	*p*-Value [b]	GMR [b,c]	95% CI [b,c]	*p*-Value [b]
Al	0.10	0.043, 0.23	0.88	0.38, 2.04	0.0003	2.50	1.33, 4.71	0.005
As	1.00	Reference	2.66 [d,e]	0.30, ∞ [d,e]	NA	4.21 [d]	0.80, 235.33 [d]	NA
Ba	0.0064	0.0026, 0.016	0.037	0.018, 0.076	0.003	2.19	1.31, 3.67	0.003
BC	4.81	1.65, 13.98	80.22	24.43, 263.43	0.0006	5.67	1.55, 20.75	0.009
Br	0.015 [f]	0.010, 0.021 [f]	0.038 [f]	0.027, 0.054 [f]	0.001 [f]	1.41 [f]	1.05, 1.90 [f]	0.03 [f]
BrC	11.32 [f]	8.99, 14.25 [f]	14.92 [f]	10.78, 20.66 [f]	0.15 [f]	1.25 [f]	0.93, 1.66 [f]	0.12 [f]
Cs	0.0045	0.0025, 0.0080	0.012	0.0068, 0.019	0.02	1.59	1.12, 2.26	0.009
Ca	0.060 [f]	0.021, 0.17 [f]	0.53 [f]	0.19, 1.49 [f]	0.006 [f]	2.59 [f]	1.22, 5.50 [f]	0.02 [f]
Ce	1.00	Reference	1.00 [d,e]	0.053, ∞ [d,e]	NA	1.21 [d]	0.034, 6.51 [d]	NA
Cl	0.083 [f]	0.035, 0.20 [f]	3.26 [f]	1.38, 7.75 [f]	<0.0001 [f]	3.86 [f]	1.58, 9.42 [f]	0.006 [f]
Cr	0.00097	0.00045, 0.0021	0.0032	0.0022, 0.0048	0.005	1.53	1.06, 2.21	0.02
Co	1.00	Reference	2.66 [d,e]	0.30, ∞ [d,e]	NA	4.21 [d]	0.80, 235.33 [d]	NA
Cu	0.0022	0.0013, 0.0038	0.012	0.0076, 0.019	<0.0001	1.66	1.01, 2.73	0.05
Fe	0.086 [f]	0.034, 0.21 [f]	0.72 [f]	0.29, 1.79 [f]	0.003 [f]	2.49 [f]	1.26, 4.94 [f]	0.01 [f]
Pb	0.0069	0.0035, 0.013	0.026	0.015, 0.045	0.002	1.49	0.93, 2.38	0.10
Mg	0.014	0.0059, 0.034	0.067	0.029, 0.15	0.01	1.90	1.03, 3.50	0.04
Mn	0.0031	0.0013, 0.0074	0.021	0.0097, 0.045	0.001	2.35	1.34, 4.13	0.003
Ni	0.0011	0.00069, 0.0018	0.0023	0.0017, 0.0031	0.01	1.28	1.01, 1.61	0.04
P	0.0042	0.0022, 0.0081	0.029	0.016, 0.052	<0.0001	2.21	1.36, 3.58	0.001
K	0.91 [f]	0.52, 1.60 [f]	2.62 [f]	1.49, 4.61 [f]	0.01 [f]	1.62 [f]	1.08, 2.42 [f]	0.02 [f]
Rb	0.0017	0.00060, 0.0051	0.0095	0.0045, 0.020	0.008	2.02	1.14, 3.57	0.02
Se	0.0023	0.0014, 0.0040	0.0037	0.0027, 0.0051	0.11	1.07	0.83, 1.39	0.59
Si	0.48 [f]	0.23, 0.96 [f]	2.32 [f]	1.15, 4.71 [f]	0.004 [f]	1.98 [f]	1.17, 3.35 [f]	0.01 [f]
Na	0.14	0.078, 0.27	0.096	0.051, 0.18	0.36	1.01	0.66, 1.55	0.96
Sr	1.00	Reference	1.00 [d,e]	0.053, ∞ [d,e]	NA	2.28 [d,e]	0.93, ∞ [d,e]	NA
S	2.21 [f]	1.69, 2.88 [f]	3.64 [f]	2.79, 4.76 [f]	0.01 [f]	1.25 [f]	1.04, 1.52 [f]	0.02 [f]
Ti	0.0063	0.0024, 0.016	0.066	0.026, 0.17	0.0005	2.76	1.38, 5.52	0.004
V	0.00077	0.00029, 0.0020	0.0048	0.0028, 0.0083	0.001	2.06	1.32, 3.20	0.002
Zn	0.035 [f]	0.021, 0.061 [f]	0.10 [f]	0.060, 0.18 [f]	0.01 [f]	1.46 [f]	0.95, 2.23 [f]	0.08 [f]

Abbreviations: Al, aluminum; As, arsenic; Ba, barium; BC, black carbon; Br, bromine; BrC, brown carbon; Cs, cesium; Ca, calcium; Ce, cerium; Cl, chlorine; Cr, chromium; Co, cobalt; CI, confidence interval; Cu, copper; GM, geometric mean; GMR, geometric mean ratio; Fe, iron; Pb, lead; Mg, magnesium; Mn, manganese; Ni, nickel; NA, not applicable; $PM_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 μm; P, phosphorus; K, potassium; Rb, rubidium; Se, selenium; Si, silicon; Na, sodium; Sr, strontium; S, sulfur; Ti, titanium; V, vanadium; and Zn, zinc. [a] The filter of one sample tore and could not be analyzed, so that home was excluded from analyses. [b] Estimated via simple (i.e., unadjusted) Tobit regression models of the natural logarithm transformed values. [c] Exponentiated regression coefficient and 95% CI (i.e., GM $PM_{2.5}$ chemical component concentration ratio for a specified change in the independent variable or $\exp(\beta) - 1$ = percent change in GM $PM_{2.5}$ chemical component concentration for a specified change in the independent variable). [d] Exact odds ratio and 95% CI; estimated via simple (i.e., unadjusted) exact unconditional logistic regression models. [e] Median unbiased estimate. [f] Estimated via simple (i.e., unadjusted) linear regression models of the natural logarithm transformed values.

Table 4. Associations between the mean of samples inside or outside homes [a] and fuel type and location at a brick kiln in Bhaktapur, Nepal (May 2019).

	Fuel Type and Location									
	LPG, Indoor		**Wood, Indoor**		**Outdoor**			**LPG, Indoor vs. Wood, Indoor**	**LPG, Indoor vs. Outdoor**	**Wood, Indoor vs. Outdoor**
$PM_{2.5}$ Chemical Component, μg/m^3	**GM ^{b}**	**95% CI ^{b}**	**GM ^{b}**	**95% CI ^{b}**	**GM ^{b}**	**95% CI ^{b}**	***p*-Value ^{b}**	***p*-Value ^{b,c}**	***p*-Value ^{b,c}**	***p*-Value ^{b,c}**
Al	0.14	0.062, 0.33	1.48	0.44, 4.98	0.37	0.074, 1.85	0.007	0.005	0.56	0.37
As	1.00	Reference	6.36 ^{d,e}	0.70, ∞ ^{d,e}	^{f}	^{f}	NA	NA	NA	NA
Ba	0.0088	0.0047, 0.016	0.062	0.028, 0.14	0.031	0.011, 0.089	0.0005	0.0004	0.11	0.56
BC	6.27	2.73, 14.41	349.04	64.46, 1,889.91	5.36	1.02, 28.21	0.0001	<0.0001	0.98	0.002
Br	0.018 ^{g}	0.013, 0.025 ^{g}	0.043 ^{g}	0.026, 0.072 ^{g}	0.013 ^{g}	0.0069, 0.025 ^{g}	0.01 ^{g}	0.02 ^{g}	0.67 ^{g}	0.02 ^{g}
BrC	12.03 ^{g}	9.09, 15.91 ^{g}	17.52 ^{g}	6.93, 44.32 ^{g}	5.54 ^{g}	3.24, 9.47 ^{g}	0.03 ^{g}	0.68 ^{g}	0.04 ^{g}	0.09 ^{g}
Cs	0.0052	0.0033, 0.0081	0.015	0.0085, 0.028	0.012	0.0055, 0.028	0.009	0.01	0.15	0.91
Ca	0.081 ^{g}	0.032, 0.21 ^{g}	1.01 ^{g}	0.25, 4.11 ^{g}	0.17 ^{g}	0.028, 1.06 ^{g}	0.02 ^{g}	0.02 ^{g}	0.72 ^{g}	0.26 ^{g}
Ce	1.00	Reference	2.20 ^{d,e}	0.12, ∞ ^{d,e}	^{f}	^{f}	NA	NA	NA	NA
Cl	0.17 ^{g}	0.072, 0.42 ^{g}	5.86 ^{g}	1.59, 21.52 ^{g}	0.071 ^{g}	0.013, 0.38 ^{g}	0.0003 ^{g}	0.0006 ^{g}	0.59 ^{g}	0.001 ^{g}
Cr	0.0013	0.00071, 0.0023	0.0039	0.0023, 0.0066	0.0034	0.0016, 0.0071	0.01	0.01	0.09	0.95
Co	1.00	Reference	6.36 ^{d,e}	0.70, ∞ ^{d,e}	12.47 ^{d,e}	1.31, ∞ ^{d,e}	NA	NA	NA	NA
Cu	0.0032	0.0019, 0.0054	0.015	0.0075, 0.030	0.0020	0.00064, 0.0064	0.0006	0.001	0.74	0.009
Fe	0.12 ^{g}	0.051, 0.28 ^{g}	1.25 ^{g}	0.35. 4.44 ^{g}	0.33 ^{g}	0.064, 1.69 ^{g}	0.02 ^{g}	0.01 ^{g}	0.48 ^{g}	0.38 ^{g}
Pb	0.0095	0.0061, 0.015	0.036	0.020, 0.064	0.014	0.0061, 0.031	0.002	0.001	0.70	0.15
Mg	0.017	0.0083, 0.036	0.11	0.040, 0.30	0.054	0.014, 0.21	0.01	0.009	0.30	0.68
Mn	0.0042	0.0022, 0.0077	0.040	0.017, 0.090	0.015	0.0048, 0.044	<0.0001	<0.0001	0.12	0.33
Ni	0.0015	0.0011, 0.0021	0.0021	0.0014, 0.0033	0.0028	0.0016, 0.0047	0.11	NA	NA	NA
P	0.0061	0.0036, 0.010	0.045	0.022, 0.092	0.017	0.0065, 0.045	<0.0001	<0.0001	0.15	0.25
K	0.98 ^{g}	0.67, 1.45 ^{g}	4.17 ^{g}	2.34, 7.43 ^{g}	0.98 ^{g}	0.46, 2.06 ^{g}	0.001 ^{g}	0.001 ^{g}	>0.99 ^{g}	0.01 ^{g}
Rb	0.0027	0.0016, 0.0046	0.018	0.011, 0.031	0.0048	0.0022, 0.010	<0.0001	<0.0001	0.46	0.01
Se	0.0031	0.0023, 0.0042	0.0032	0.0022, 0.0048	0.0039	0.0025, 0.0062	0.71	NA	NA	NA
Si	0.60 ^{g}	0.30, 1.20 ^{g}	3.58 ^{g}	1.28, 9.97 ^{g}	1.23 ^{g}	0.33, 4.62 ^{g}	0.03 ^{g}	0.02 ^{g}	0.58 ^{g}	0.39 ^{g}
Na	0.13	0.079, 0.21	0.097	0.046, 0.20	0.21	0.083, 0.54	0.44	NA	NA	NA
Sr	1.00	Reference	2.20 ^{d,e}	0.12, ∞ ^{d,e}	3.67 ^{d,e}	0.19, ∞ ^{d,e}	NA	NA	NA	NA
S	2.35 ^{g}	1.80, 3.06 ^{g}	4.29 ^{g}	2.90, 6.35 ^{g}	2.45 ^{g}	1.47, 4.06 ^{g}	0.04 ^{g}	0.04 ^{g}	0.99 ^{g}	0.18 ^{g}
Ti	0.0088	0.0037, 0.021	0.13	0.036, 0.46	0.028	0.0052, 0.15	0.003	0.002	0.46	0.34
V	0.0013	0.00079, 0.0022	0.0075	0.0043, 0.013	0.0035	0.0017, 0.0075	<0.0001	<0.0001	0.08	0.25
Zn	0.039 ^{g}	0.026, 0.060 ^{g}	0.15 ^{g}	0.084, 0.28 ^{g}	0.053 ^{g}	0.024, 0.12 ^{g}	0.004 ^{g}	0.003 ^{g}	0.75 ^{g}	0.09 ^{g}

Abbreviations: Al, aluminum; As, arsenic; Ba, barium; BC, black carbon; Br, bromine; BrC, brown carbon; Cs, cesium; Ca, calcium; Ce, cerium; Cl, chlorine; Cr, chromium; Co, cobalt; CI, confidence interval; Cu, copper; GM, geometric mean; Fe, iron; Pb, lead; LPG, liquefied petroleum gas; Mg, magnesium; Mn, manganese; Ni, nickel; NA, not applicable; $PM_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 μm; P, phosphorus; K, potassium; Rb, rubidium; Se, selenium; Si, silicon; Na, sodium; Sr, strontium; S, sulfur; Ti, titanium; V, vanadium; and Zn, zinc. [a] The filter of one inside sample tore and could not be analyzed, so that home was excluded from analyses. Of the remaining 19 samples, 16 were from inside and three were from outside the homes. [b] Estimated via simple (i.e., unadjusted) Tobit regression models of the natural logarithm transformed values. [c] Used the Tukey-Kramer (for Tobit regression models) or Tukey (for linear regression models) methods to adjust for multiple comparisons. [d] Exact odds ratio and 95% CI; estimated via simple (i.e., unadjusted) exact unconditional logistic regression models. [e] Median unbiased estimate. [f] Degenerate; unable to estimate. [g] Estimated via simple (i.e., unadjusted) linear regression models of the natural logarithm transformed values.

3.4. Adjusted Associations between Characteristics of Homes of Brick Workers and $PM_{2.5}$ Chemical Component Concentrations

Smokers in the home was most consistently significantly associated with $PM_{2.5}$ Cl and $PM_{2.5}$ Cu when some combination of home area, smokers in the home or number of smokers in the home, and fuel type were included as independent variables in the multivariable linear regression models (Supplementary Materials, Table S3). Smokers in the home was significantly associated with concentrations of six (24%) $PM_{2.5}$ chemical components, and fuel type was significantly associated with concentrations of nine (36%)

$PM_{2.5}$ chemical components when smokers in the home and fuel type were included as independent variables in the multivariable linear or Tobit regression models (Table 5). The number of smokers in the home was not significantly associated with concentrations of any $PM_{2.5}$ chemical component, but fuel type was significantly associated with concentrations of 14 (56%) $PM_{2.5}$ chemical components when the number of smokers in the home and fuel type were included as independent variables in the multivariable linear or Tobit regression models (Table 6).

Table 5. Associations between the mean of samples inside homes [a] and smokers in home and fuel type, mutually adjusted for each other at a brick kiln in Bhaktapur, Nepal (May 2019).

	Smokers in Home	**Fuel Type**
$PM_{2.5}$ Chemical Component	***p*-Value [b]**	***p*-Value [b]**
Al	0.08	0.09
Ba	0.23	0.03
BC	0.22	0.002
Br	0.04 [c]	0.30 [c]
BrC	0.30 [c]	0.55 [c]
Cs	0.39	0.12
Ca	0.20 [c]	0.08 [c]
Cl	0.002 [c]	0.06 [c]
Cr	0.10	0.20
Cu	0.003	0.22
Fe	0.14 [c]	0.09 [c]
Pb	0.13	0.10
Mg	0.39	0.09
Mn	0.22	0.008
Ni	0.02	0.62
P	0.02	0.02
K	0.50 [c]	0.02 [c]
Rb	0.68	0.002
Se	0.05	0.24
Si	0.16 [c]	0.09 [c]
Na	0.50	0.99
S	0.32 [c]	0.09 [c]
Ti	0.12	0.04
V	0.07	0.004
Zn	0.37 [c]	0.03 [c]

Abbreviations: Al, aluminum; Ba, barium; BC, black carbon; Br, bromine; BrC, brown carbon; Cs, cesium; Ca, calcium; Cl, chlorine; Cr, chromium; Cu, copper; Fe, iron; Pb, lead; Mg, magnesium; Mn, manganese; Ni, nickel; $PM_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 μm; P, phosphorus; K, potassium; Rb, rubidium; Se, selenium; Si, silicon; Na, sodium; S, sulfur; Ti, titanium; V, vanadium; and Zn, zinc. [a] The filter of one sample tore and could not be analyzed, so that home was excluded from analyses. [b] Estimated via multivariable Tobit regression models of the natural logarithm transformed values adjusted for smokers in the home and fuel type. [c] Estimated via multivariable linear regression models of the natural logarithm transformed values adjusted for smokers in the home and fuel type.

Table 6. Associations between the mean of samples inside homes [a] and the number of smokers in the home and fuel type, mutually adjusted for each other at a brick kiln in Bhaktapur, Nepal (May 2019).

	Number of Smokers in Home	**Fuel Type**
$PM_{2.5}$ Chemical Component	***p*-Value [b]**	***p*-Value [b]**
Al	0.35	0.04
Ba	0.30	0.02
BC	0.72	0.002
Br	0.48 [c]	0.11 [c]
BrC	0.30 [c]	0.99 [c]

Table 6. *Cont.*

	Number of Smokers in Home	Fuel Type
$PM_{2.5}$ Chemical Component	p-Value [b]	p-Value [b]
Cs	0.30	0.14
Ca	0.46 [c]	0.06 [c]
Cl	0.32 [c]	0.02 [c]
Cr	0.50	0.07
Cu	0.98	0.01
Fe	0.39 [c]	0.06 [c]
Pb	0.65	0.004
Mg	0.77	0.04
Mn	0.39	0.005
Ni	0.09	0.82
P	0.27	0.006
K	0.75 [c]	0.01 [c]
Rb	0.92	0.0007
Se	0.57	0.82
Si	0.43 [c]	0.06 [c]
Na	0.51	0.37
S	0.51 [c]	0.07 [c]
Ti	0.39	0.02
V	0.20	0.002
Zn	0.70 [c]	0.008 [c]

Abbreviations: Al, aluminum; Ba, barium; BC, black carbon; Br, bromine; BrC, brown carbon; Cs, cesium; Ca, calcium; Cl, chlorine; Cr, chromium; Cu, copper; Fe, iron; Pb, lead; Mg, magnesium; Mn, manganese; Ni, nickel; $PM_{2.5}$, particulate matter with an aerodynamic diameter less than 2.5 µm; P, phosphorus; K, potassium; Rb, rubidium; Se, selenium; Si, silicon; Na, sodium; S, sulfur; Ti, titanium; V, vanadium; and Zn, zinc. [a] The filter of one sample tore and could not be analyzed, so that home was excluded from analyses. [b] Estimated via multivariable Tobit regression models of the natural logarithm transformed values adjusted for the number of smokers in the home and fuel type. [c] Estimated via multivariable linear regression models of the natural logarithm transformed values adjusted for the number of smokers in the home and fuel type.

3.5. Sensitivity Analyses

The results were almost identical when we repeated analyses using home volume instead of home area (not shown), with the one exception being that home volume was not significantly associated with concentrations of $PM_{2.5}$ Cl (8.52–16.00 m^3: GM = 1.23; 95% CI: 0.26, 5.94 $\mu g/m^3$; and >16.00–53.44 m^3: GM = 0.22; 95% CI: 0.046, 1.06 $\mu g/m^3$; $p = 0.12$). In other words, home volume was significantly associated with concentrations of $PM_{2.5}$ Cu (8.52–16.00 m^3: GM = 0.0086; 95% CI: 0.0043, 0.017 $\mu g/m^3$; and >16.00–53.44 m^3: GM = 0.0030; 95% CI: 0.0014, 0.0063 $\mu g/m^3$; $p = 0.04$), but not with concentrations of any other $PM_{2.5}$ chemical component (not shown).

4. Discussion

This research was conducted as a follow-up to a previous study we conducted in 2018 [18]. In our previous study, we collected $PM_{2.5}$ samples in on-site brick workers' homes in Bhaktapur, Nepal, but the sampling time was limited to approximately seven hours during the middle of the day when most workers were not at home. Thus, we were not able to measure $PM_{2.5}$ generated during cooking and other household activities during non-working hours. The longer sampling time (approximately 21 h in each home) used in the current study allowed us to characterize $PM_{2.5}$ constituents across both working and non-working hours. Using the seven-hour samples in our previous study, we found no difference in the chemical composition of indoor vs. outdoor air, except for Cl, which was higher indoors. Our previous study also found that the primary fuel used for cooking was significantly associated with only two $PM_{2.5}$ chemical components, Cl and K, which were both higher in wood fuel homes. In contrast, in this study we found significant differences for 22 chemical components based on cooking fuel type and location (LPG, indoor vs. wood, indoor vs. outdoor). Pairwise comparisons indicated fuel type was the primary source of these significant differences. We attributed these differences in results among studies to non-working hour activities in the home that were not captured in our previous study, but were captured in our current study.

The major elemental aerosol-phase tracers of wood smoke are Cl and K, both of which are commonly found in $PM_{2.5}$ generated from wood combustion [41]. Like our previous study, we found significantly higher levels of both elements in homes where wood fires were used for cooking. For wood fire homes, the indoor Cl level averaged 5.86 $\mu g/m^3$, which was approximately 34 times the average level in LPG homes (0.17 $\mu g/m^3$). Similarly, K levels in wood fire homes (4.17 $\mu g/m^3$) were approximately four times the levels in LPG homes (0.98 $\mu g/m^3$). Furthermore, our results showed significant differences in Cl and K levels between wood fire homes and outdoor air, but not between LPG homes and outdoor air, suggesting the high levels of Cl and K in our study originated from cooking indoors over wood fires. Our findings are consistent with previous studies conducted in homes in West Africa and India. In higher-income homes in Accra, Ghana, where residents tend to cook indoors with LPG cookstoves, Zhou et al. reported average indoor Cl and K levels of 0.34 and 1.08 $\mu g/m^3$, respectively [42]. By comparison, average Cl and K levels in enclosed cookhouses using firewood in The Gambia were 7.90 and 10.75 $\mu g/m^3$, respectively [42]. In unventilated, low-income homes in India where solid biomass fuels were used for cooking, annual Cl and K levels averaged 5.8 and 7.6 $\mu g/m^3$, respectively [43].

Among the 35 analytes, BC accounted for the highest concentration in wood fire homes (349.04 $\mu g/m^3$), where levels were 56 and 65 times the levels in LPG homes and outdoor air, respectively. BC is released into the air as a result of incomplete combustion of fuels, and prolonged or extreme exposure is associated with increased morbidity and mortality, primarily from cardiac and respiratory illnesses [15]. When BC acts as a carrier for polycyclic aromatic hydrocarbons, it is linked to adverse health effects, including cancer and severe immune, reproductive, and pulmonary damage [15,44]. Several additional elemental species identified in our study were previously shown to be associated with burning wood. For example, we found concentrations of Al, Ca, magnesium (Mg), phosphorus (P), and Si were significantly higher in homes with wood cooking fires than in homes with LPG cookstoves. All of these elements were shown in previous studies to be associated with high temperature burning of wood or wood pellets in stoves [23,45].

Respiratory illnesses are common among brick workers in Nepal [21], and occupational exposures likely play an important role in this finding [20]. However, previous studies of urban ambient $PM_{2.5}$ constituents found that several metals and other elements were associated with respiratory disease in adults and children [10,12] at much lower concentrations than those found in our study. For example, Bell et al. found associations between respiratory hospital admissions in adults $\geq$65 years of age and $PM_{2.5}$ constituents Al, Ca, Cl, BC, Ni, Si, Ti, and V [10]. In our study, all of these constituents, with the exception of Ni and V, were found in higher concentrations than those reported by Bell et al. in all sampled locations (wood indoor, LPG indoor, and outdoor air), and V concentrations in our study were higher in wood fire homes. Ostro et al. found associations between respiratory hospital admissions in children and concentrations of Cu, Fe, K, Si, and Zn in ambient air [12]. Again, we found each of these constituents in our samples, and in most cases at higher concentrations than those reported by Ostro et al. Differences in concentrations were most pronounced in wood fire homes, where element concentrations ranged from 1.9–35 times the ambient concentrations reported by Ostro et al. We propose that repeated exposure to the high concentrations of metals and other elements in both indoor and outdoor air may contribute significantly to the respiratory symptoms seen among brick workers in Nepal.

One of the most noticeable differences between this study and our previous one was the number of chemical components that had significantly higher concentrations in homes with smokers. Depending on the tobacco source, cigarette smoke contains varying levels of several metals that are associated with deleterious health effects, including Al, arsenic (As), Ba, beryllium (Be), cadmium (Cd), cobalt (Co), chromium (Cr), Cu, Fe, lead (Pb), manganese (Mn), mercury (Hg), Ni, selenium (Se), Si, V, and Zn [46,47]. Our current study found these metals in higher concentrations in homes with smokers compared to homes with non-smokers, with the exception of Cd, which had all sample concentrations below the LDL, and Be and Hg, which we did not test for. Toxicologically, these metals are associated with allergic sensitization and inflammation, COPD, cancer, asthma, immune system suppression [47], vascular endothelium damage, and the development of atherosclerosis [46].

In our previous study, we discussed concerns about small, overcrowded housing among brick workers in Nepal, and specifically regarding the potential for indoor pollution to concentrate in smaller, poorly ventilated homes [18]. The finding that home area was significantly associated with $PM_{2.5}$ Cl and Cu appears to support this concern. In the cases of both Cl and Cu, smaller homes (i.e., 5.41–9.50 m^2) had the highest concentrations (Supplementary Materials, Table S1). Although not statistically significant, smaller homes also had the highest GM concentrations for 22 other chemical components. Smaller home area appears to contribute to a build-up in air pollution concentrations. The small sample size and reduced statistical power in this study may have contributed to our inability to detect significant associations between home area and concentrations of $PM_{2.5}$ constituents for elements other than Cl and Cu.

We used multivariable linear or Tobit regression models that included two or three characteristics of homes at the brick kilns (i.e., smoking, fuel type, and home area) as independent variables to determine whether significant associations between these characteristics and concentrations of $PM_{2.5}$ chemical components, found using simple (unadjusted) regression models, remained statistically significant when we adjusted for the other characteristic(s). As stated previously, Cu, Ni, and Se were previously found in cigarette smoke [46,47] and all three $PM_{2.5}$ chemical components were significantly associated with smokers in the home in our study when we adjusted for fuel type. In addition, Cu was significantly associated with smokers in the home when we adjusted for home area and fuel type. Al, BC, Cl, Mg, P, and K were previously found in wood smoke [15,23,41,45] and all six $PM_{2.5}$ chemical components were significantly associated with fuel type in our study when we adjusted for smokers in the home and/or number of smokers in the home. However, Cl was not significantly associated with fuel type when we adjusted for home area and smokers in the home or number of smokers in the home. Cl was instead significantly associated with smokers in the home when we adjusted for home area and fuel type. Ba, Pb, Mn, V, and Zn were previously found in cigarette smoke [46,47], but none of these $PM_{2.5}$ chemical components were significantly associated with smokers in the home or number of smokers in the home in our study when we adjusted for fuel type. All five $PM_{2.5}$ chemical components were instead significantly associated with fuel type when we adjusted for smokers in the home and/or number of smokers in the home. The reasons for these discrepancies between our results and those of previous studies are unknown, but our small sample size and the fact that all five homes that used wood for fuel also had smokers in the home may have contributed.

Although we did not have a sufficient sample size to conduct principal component analysis in this study, we can make some conjecture about possible pollution sources. Of the 29 analytes that had at least one sample concentration above the LDL, only one (BrC) was significantly different between LPG homes and outdoor air. This finding may be explained by stir-fry cooking within the home [48], or possibly by activities such as burning candles or smoking indoors during non-working hours. Non-significant differences in the remaining 28 analytes may be largely explained by infiltration of ambient air pollution through gaps in brick workers' homes, as discussed previously [18,19]. There are currently over 100 operating brick kilns in the Kathmandu Valley, most of which are coal fired [17,49,50]. Several analytes from our samples are known to originate from coal burning, such as Al, As, Ba, Ca, Fe, K, Mg, Mn, P, Se, Si, and Ti, depending on the source of the coal [51]. In addition, the kiln from which our samples were collected is located near the Araniko Highway, a major roadway through Bhaktapur. Vehicle exhaust is a source of several metals that we found in ambient air, as well as in participant homes, including Cr, Cu, Fe, Ni, Pb, and Zn [52–54]. Tire fading and brake wear may be responsible for Zn and Cd, and Cu and Zn, respectively [52,55], although Cd concentrations were below the LDL for all samples in our study.

One limitation in understanding the contribution of wood burning to elemental composition in our study is that we did not measure burn temperature, which greatly affects the chemical composition of particles [24]. In future studies, we may also consider using the EC/OC ratio or measuring methyl chloride levels as more definitive markers of wood smoke in our study homes, as well as looking more closely at the bioavailability of PM-bound metals to understand the toxicological properties of $PM_{2.5}$ in brick workers' homes. We were unable to obtain measurements for BrC for four samples because the amount of BC on the filters surpassed the UDL, which rendered the optical transmittance method unfeasible. This study was also limited because samples were obtained from homes at a single brick kiln and we had a relatively small sample size. A larger sample size would have allowed for the use of principal component analysis or related methods, such as positive matrix factorization, which was used in other studies [42], to determine the sources of pollution. Other limitations of this study (e.g., unmeasured confounding by temporal factors, lack of health data, etc.) were discussed previously [19].

5. Conclusions

Based on the findings of this and other studies [18,19], we suggest a multi-faceted approach is needed to protect brick workers in the Kathmandu Valley from the adverse health effects associated with poor air quality. The atmospheric pressure, wind direction and velocity, humidity, and the bowl-shaped topography of Kathmandu Valley add to the air pollution problems [56]. As air pollution remains a major issue, it is of paramount importance to educate the general population regarding the detrimental effects of air pollution and preventative measures to inhibit extreme outcomes [57]. The government of Nepal has to take primary responsibility to address the consequences of this problem by developing policies and action plans to reduce ambient air pollution and, ultimately, its consequences [57]. As the primary source of indoor air pollution in Nepal is the burning of solid fuels for cooking, improved stoves, smoke hoods, vented or chimney stoves, and clean fuel replacements would reduce the disease burden due to indoor air pollution exposure [58]. Considering 50% of homes in this study had at least one smoker, and that smoking is a significant predictor of respiratory illness among brick workers in Nepal [21], future interventions to improve indoor air quality in brick workers' homes should also include smoking cessation programs [59,60].

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/atmos12070911/s1, Table S1: Associations between the mean of samples inside homes and home area and number of people in home at a brick kiln in Bhaktapur, Nepal, May 2019, Table S2: Associations between the mean of samples inside homes and occupant density and number of children in home at a brick kiln in Bhaktapur, Nepal, May 2019, and Table S3: Associations between the mean of samples inside homes and home area, smokers in home, number of smokers in home, and fuel type mutually adjusted for each other at a brick kiln in Bhaktapur, Nepal, May 2019.

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