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Personal and Indoor $PM_{2.5}$ Exposure from Burning Solid Fuels in Vented and Unvented Stoves in a Rural Region of China with a High Incidence of Lung Cancer

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Supporting Information

ABSTRACT: The combustion of biomass and coal is the dominant source of household air pollution (HAP) in China, and contributes significantly to the total burden of disease in the Chinese population. To characterize HAP exposure related to solid fuel use and ventilation patterns, an exposure assessment study of 163 nonsmoking female heads of households enrolled from 30 villages was conducted in Xuanwei and Fuyuan, two neighboring rural counties with high incidence of lung cancer due to the burning of smoky coal (a bituminous coal, which in health evaluations is usually compared to smokeless coal—an anthracite coal available in some parts of the area). Personal and indoor 24-h $PM_{2.5}$ samples were collected over two consecutive days in each household, with approximately one-third of measurements retaken in a second season. The overall geometric means (GM) of personal $PM_{2.5}$ concentrations in Xuanwei and Fuyuan were 166 [Geometric Standard Deviation (GSD):2.0] and 146 (GSD:1.9) $\mu g/m^3$, respectively, which were similar to the indoor $PM_{2.5}$ air concentrations [GM(GSD):162 (2.1) and 136 (2.0) $\mu g/m^3$, respectively]. Personal $PM_{2.5}$ was moderately highly correlated with indoor $PM_{2.5}$ (Spearman $r = 0.70$, $p < 0.0001$). Burning wood or plant materials (tobacco stems, corncobs etc.) resulted in the highest personal $PM_{2.5}$ concentrations (GM:289 and 225 $\mu g/m^3$, respectively), followed by smoky coal, and smokeless coal (GM:148 and 115 $\mu g/m^3$, respectively). $PM_{2.5}$ levels of vented stoves were 34−80% lower than unvented stoves and firepits across fuel types. Mixed effect models indicated that fuel type, ventilation, number of windows, season, and burning time per stove were the main factors related to personal $PM_{2.5}$ exposure. Lower $PM_{2.5}$ among vented stoves compared with unvented stoves and firepits is of interest as it parallels the observation of reduced risks of malignant and nonmalignant lung diseases in the region.

INTRODUCTION

More than 60% of the population in China is rural. Nearly all of this population use biomass and coal fuels for their day to day cooking and heating, the combustion of which is the dominant source of household air pollution (HAP) in China and contributes significantly to the total burden of ill health in the Chinese population.[1] Particulate matter with an aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) is one of the main pollutants in solid fuel smoke and is closely associated with many of the adverse health effects associated with HAP.[2−9]

Xuanwei and Fuyuan, two neighboring counties in Yunnan province, China have a mostly rural population, and have increased rates of nonmalignant and malignant lung disease associated with HAP from solid fuel combustion.[6,10] In particular, the lung cancer rates in this region are among China's highest in both males and females regardless of smoking status.[11] Previous epidemiological studies have shown that the excess lung cancer risk could be mainly attributed to the domestic combustion of "smoky" coal in poorly ventilated

Received: October 21, 2013
Revised: June 26, 2014
Accepted: July 8, 2014
Published: July 8, 2014

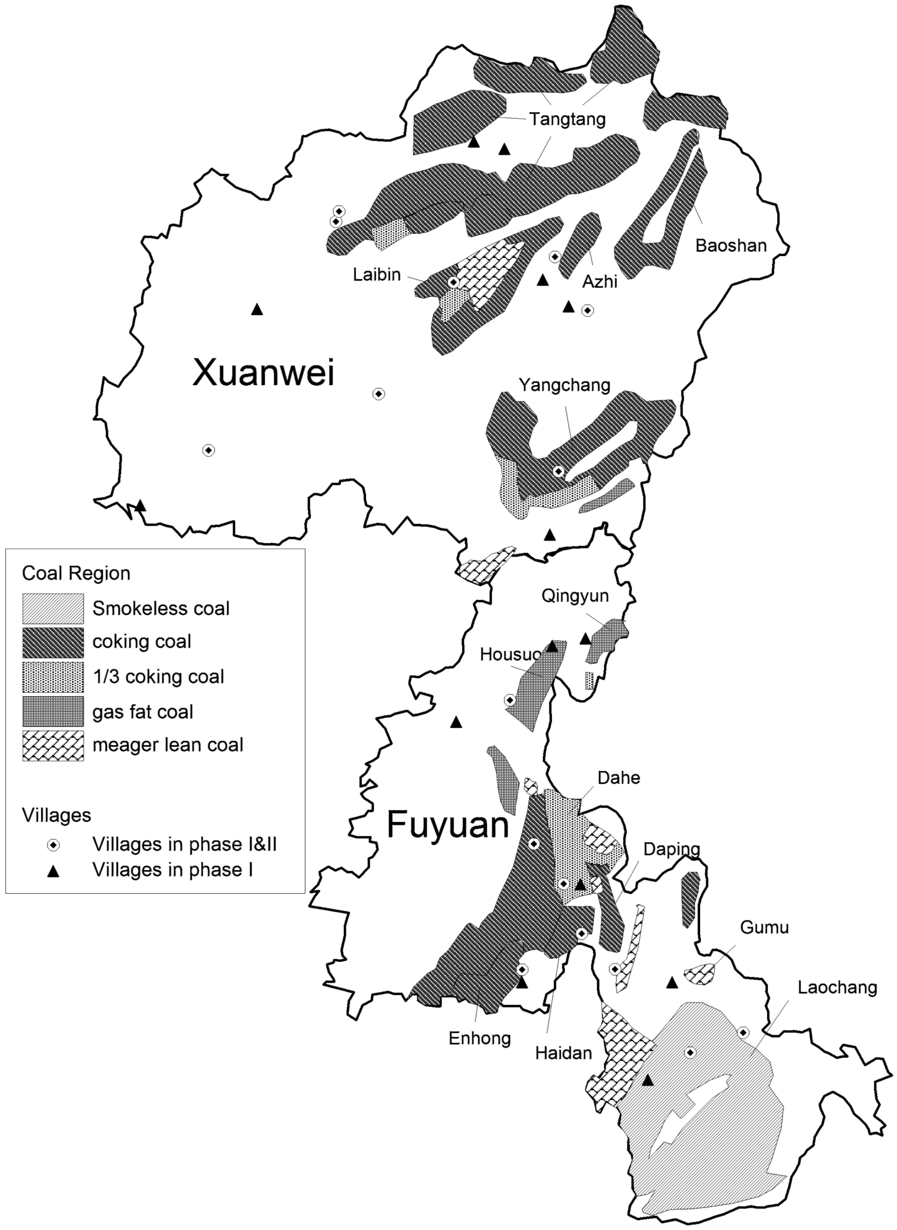

Figure 1. Villages selected for the exposure survey and coal mines in Xuanwei and Fuyuan counties. Classification of coal regions based on the State Standard of China Coal Classification (GB5751−86); 1/3 coking, coking, gas fat, and meager lean coals are subtypes of smoky coal.

homes for heating and cooking.[11−13] Coal is the major domestic fuel in this region and depending upon the source (residents typically purchased coal from their nearest mines) is either referred to as smoky or smokeless coal, terms which relate to the amount of visible smoke produced on combustion. Smoky coal is further divided into subtypes based upon the underlying geochemical properties and geographic location.[14,15] These subtypes may have varying carcinogenic potentials as a wide variation in lung cancer risk (up to 25-fold) has been observed between geographic locations in Xuanwei.[16] Additionally, stove improvement programs have resulted in a reduction in HAP and a decreased burden of nonmalignant and malignant lung disease in this region,[12,17,18] thus suggesting that HAP is important in the etiology of malignant and nonmalignant respiratory disease in the study area.

There have been few systematic indoor air quality studies in Xuanwei and Fuyuan to date, and these have mainly focused on the quantification of airborne benzo[a]pyrene (BaP).[13,19] Moreover, these studies focused on indoor air measurements, and it is unclear how these relate to personal exposure. This work presents our findings from personal and indoor $PM_{2.5}$ measurements collected from 30 villages throughout the Xuanwei and Fuyuan counties. Particular attention will be paid to differences between solid fuels (especially differences between smoky and smokeless coal), differences between stove designs (e.g., vented and unvented), and variation between smoky coal subtypes. This study also provides an opportunity to characterize the relationship between indoor and personal measurements, which may have an influence on costs, participation, and other logistical concerns for long-term studies[18,20] and provides information on the efficiency of stove improvement programs.

■ MATERIALS AND METHODS

Study Design. This exposure assessment study was designed to provide a comprehensive characterization of air contaminants and exposures related to the use of solid fuels for cooking and heating, coordinating with an ongoing large case-control study of lung cancer, and a cross-sectional molecular epidemiology study of lung cancer in the same area.

A total of 30 villages were selected to represent the major geographical regions in Xuanwei and Fuyuan (Figure 1). In each selected village, up to 5 households were selected according to the following criteria, which was intended to allow for eventual comparability to the larger case-control study: (i) having a stove that used solid fuel; (ii) the residence was more than 10 years old; (iii) use of the same cooking/heating equipment for the past 5 years; and (iv) presence of a nonsmoking healthy female aged 20−80 who was primarily responsible for cooking. The exposure survey was carried out in two phases. Phase I was carried out from August 2008 to February 2009 with all 30 villages visited and 148 subjects enrolled. Phase II was carried out between March and June of 2009. Sixteen of the initial 30 villages, chosen to represent the geographical spread, fuel use, and stove type distribution to phase I were revisited (Figure 1). In the second phase, 15 new subjects were enrolled and 53 of the enrollees from phase I were revisited allowing estimation of seasonal effects. Each sampling phase consisted of two sequential 24-h air measurements during which indoor and personal measurements were taken.

$PM_{2.5}$ Measurement. $PM_{2.5}$ measurements for 24-h were collected on preweighed 37 mm Teflon filters using a cyclone with an aerodynamic cutoff of 2.5 μm (model BGI, GK 2.05SH) at a flow rate of 3.5 L/min ($\pm$20%). Pumps were calibrated prior to all measurements and flow rates were recorded pre and post sampling. Data were not accepted for further analysis if the post sampling flow was not within 10% of the presampling flow. For personal measurements, the pump was packed in a hip bag and the cyclone was attached near the breathing zone. At night, the sampling bag was put next to the subject's bed.

Most houses had a single living/cooking area. Measurements of 24-h indoor $PM_{2.5}$ were collected in the main living area using the same methods as described for the personal measurements. The samplers were placed at least 0.25m from the walls and between 1 and 2m from the stove. Placement varied somewhat because of limited available space in some households. If a subject had a separate room with an additional stove for cooking or heating, then a second stationary measurement was taken (this represents 6% of indoor measurements).

Exposed filters were packed individually in Petri slides sealed in zipped amber plastic bags and stored at −80 °C before postweighing. Particulate mass was assessed by pre- and postweighing of the filters in an environmentally controlled weighing room using a microbalance at 1 μg accuracy. Each filter was pre- and postweighed in duplicate. If the duplicate measurements differed by more than 5 μg, then the filter was reweighed. Weights were divided by the volume of air drawn through the filters to calculate $PM_{2.5}$ concentrations ($\mu g/m^3$).

For quality control purposes, approximately 10% of households were randomly selected to have duplicate $PM_{2.5}$ measurements collected. The coefficient of variation of this quality control was 13%, based on 26 pairs of collocated indoor $PM_{2.5}$ samples.

Household Interview and Measurement. Household interviews were conducted by two trained interviewers. House dimensions were recorded and a sketch of each household was made detailing the position and dimensions of stoves, windows, chimneys, doors, and stairways. Subjects' activities during the sampling periods were recorded by administering a short activity questionnaire. The activity survey included information on cooking activities, heating practices, fuel usage, outdoor activities, and sleeping habits. Stove details were recorded with a particular focus upon the ventilation aspect of their design. Traditionally, fuel has been burned in unvented firepits, but in recent decades a variety of stove types have been utilized throughout the area, not all of which include a functioning chimney. The major stove designs encountered were as follows: vented stoves, unvented stoves, firepits, portable stoves (a stove design intended to be lit outdoors and then carried indoors for use) and a combination of these stove designs. The other major sources of ventilation recorded were the numbers of doors, windows, and the presence or absence of a stairway in the main cooking area.

Fuel types were reported by respondents. Coal was generally reported as either "smoky" or "smokeless". Coal types were confirmed by petrochemical analysis of collected coal samples and in 11 cases the classification of coal type was changed to match petrochemical analysis (7 samples were reclassified as smoky coal and 4 samples were reclassified as smokeless coal).[14] Other categories identified were as follows: wood, plant products (which included the burning of corn cobs and tobacco stems−sometimes also in combination with wood), "mixed" coal (which represented the burning of manufactured coal briquettes and combinations of briquettes, smoky and smokeless coal), and "mixed" fuel (which represented the burning of combinations of wood, plant materials and coal). The coal mines supplying coal were ascertained at interviews during household visits. Smoky coal burned during the exposure measurements came from 5 coal mines in Xuanwei and 8 mines in Fuyuan. Smoky coal was divided into subtypes according to the parameters of the State Standard of China Coal Classification (GB5751-86).[15] These smoky coal subtypes, based upon the parameters of volatile matter on a dry ash free basis and caking index, are referred to as coking, 1/3 coking, gas fat, and meager lean coals.

On each measuring day, weather parameters including outside temperature, humidity, rainfall, wind speed, and direction were monitored by a portable weather station set up in the center of each village.

Statistical Analysis. Normal probability plots indicated that exposures could best be described by a log-normal distribution. Therefore, natural logarithms of exposure concentrations were used in the statistical analyses. Exposure levels were summarized as arithmetic means (AM), geometric means (GM), and geometric standard deviations (GSD). Analysis of variance (ANOVA) testing was performed to test for differences in $PM_{2.5}$ exposure between differing stove and fuel configurations and for variation within the designated smoky coal subtypes. Tukey Honestly Significant Difference (HSD) testing was performed to assess pairwise differences within each combination of two levels of the various fuel and stove configurations.

A linear mixed effect model was constructed to identify variables that contributed to personal $PM_{2.5}$ exposure. Subjects

Table 1. Characteristics of the Study Population in Xuanwei and Fuyuan

	phase I		phase II		all
	Xuanwei	Fuyuan	Xuanwei	Fuyuan	
subjects, *n*	74	74	31	37	163[a]
villages, *n*	15	15	8	8	30
age (in 2009), mean ± SD	54.0 ± 14.9	56.7 ± 13.7	62.0 ± 11.3	58.9 ± 12.2	56.0 ± 14.4
stove type, *n*(%)					
vented stove	34(45.9)	19(25.7)	12(38.7)	10(27.0)	75(34.7)
high stove with chimney	*13(17.6)*	*5(6.8)*	*6(19.4)*	*1(2.7)*	*25(11.6)*
low stove with chimney	*8(10.8)*	*11(14.9)*	*2(6.5)*	*6(16.2)*	*27(12.5)*
multiple stoves with chimneys	*13(17.6)*	*3(4.1)*	*4(12.9)*	*3(8.1)*	*23(10.6)*
unvented stove	4(5.4)	12(16.2)	0(0.0)	11(29.7)	27(12.5)
high stove without chimney	*0(0.0)*	*6(8.1)*	*0(0.0)*	*2(5.4)*	*8(3.7)*
low stove without chimney	*0(0.0)*	*0(0.0)*	*0(0.0)*	*1(2.7)*	*1(0.5)*
multiple stoves without any chimney	*4(5.4)*	*6(8.1)*	*0(0.0)*	*8(21.6)*	*18(8.3)*
portable stove	2(2.7)	19(25.7)	1(3.2)	8(21.6)	30(13.9)
firepit	3(4.1)	7(9.5)	3(9.7)	3(8.1)	16(7.4)
mixed ventilation stoves[b]	30(40.5)	13(17.6)	13(41.9)	5(13.5)	61(28.2)
unknown ventilation stove	1(1.4)	4(5.4)	2(6.5)	0(0.0)	7(3.2)
solid fuel type, n(%)					
smoky coal	42(56.8)	32(43.2)	19(61.3)	6(16.2)	99(45.8)
smokeless coal	0(0.0)	13(17.6)	1(3.2)	5(13.5)	19(8.8)
"mixed" coal[c]	9(12.2)	5(6.8)	1(3.2)	4(10.8)	19(8.8)
wood	3(4.1)	1(1.4)	2(6.5)	6(16.2)	11(5.1)
plant materials[d]	4(5.4)	3(4.1)	1(3.2)	0(0.0)	9(4.2)
"mixed" fuel[e]	16(21.6)	18(24.3)	6(19.4)	16(43.2)	56(25.9)
unknown	0(0.0)	2(2.7)	1(3.2)	0(0.0)	3(1.4)
median length of stove operation, hours per day	4	3.3	8	13	5.1

[a]There were 216 visits to the households in total: of the 148 subjects visited in phase I, 53 were revisited the second time, and 15 new subjects were enrolled in phase II. [b]Refers to the use of vented stove and unvented stove/portable stove simultaneously. [c]Refers to the use of combinations of smoky and smokeless coal and also to the use of prepared coal briquettes. [d]Plant materials include combinations of wood, tobacco stem and corncob. [e]Refers to combinations of wood, plant materials and coal.

and villages were assigned as random effects with a scalar (variance component) covariance structure. Multiple variables were considered for inclusion as fixed effects including: stove design, fuel type (including both broad fuel categories and the inclusion of smoky coal subtypes), fuel source, weight of fuel used, meteorological conditions, room size, number of owned stoves, number of doors, windows and the presence or absence of a stairway in the main cooking room, hours of using stoves, and the season during which measurements were taken. A full list of the 32 considered variables is available in Supporting Information (SI) Table S1. The variables selected for inclusion in the final model were those which best contributed to the prediction of $PM_{2.5}$ measurements and contributed to the lowest Akaike information criterion (AIC) score. The model can be summarized using the following formula:

$$y_{\mathrm{ijf}} = \mu + \beta_1 x_1 + \beta_2 x_2 ... \beta_n x_n + b\mathrm{I}_{\mathrm{i}} + b\mathrm{J}_{\mathrm{ij}} + \varepsilon_{\mathrm{ijf}}$$

where y_{ijf} represents the natural log transformed value of personal $PM_{2.5}$ exposure being modeled for village i, person j on day f; μ represents the intercept value (i.e., the log transformed $PM_{2.5}$ value for the reference group); β_1 through to β_n represent the fixed effect variable coefficients for variables x_1 through x_n; bI_i represents the random effect coefficient for village i, while bJ_{ij} represents the random effect coefficient for subject j from village i; and ε_{ijf} represents the error for subject j in village i on day f.

Spearman correlation was calculated between personal and indoor $PM_{2.5}$ measurements collected on the same day and a linear mixed effect model was constructed to identify which variables best explained any differences in the association between personal and indoor $PM_{2.5}$ measurements.

RESULTS

A total of 163 subjects participated in the exposure survey and 216 household visits were conducted. An overview of the characteristics of the study population is available in Table 1. Smoky coal was the main fuel type used during the measurement in both counties (45.8%), followed by "mixed" fuels (25.9%), and smokeless coal (8.8%). The most commonly used stove design was vented stoves (34.7%) followed by the use of multiple stoves with differing ventilation designs [which we refer to as "mixed ventilation" (28.2%)]. The average age of the subjects in the exposure survey was 56 years. The distribution of the villages visited is shown in Figure 1.

The overall GM of personal $PM_{2.5}$ concentrations in Xuanwei and Fuyuan were 166 (GSD:2.0) and 146 (GSD:1.9) $\mu g/m^3$, respectively. These levels were similar to the overall indoor $PM_{2.5}$ concentration [GM(GSD):162 (2.1) and 136 (2.0) $\mu g/m^3$ for Xuanwei and Fuyuan, respectively]. Personal and indoor $PM_{2.5}$ measurements correlated well, with Spearman correlation analysis indicating a moderately high coefficient between personal and indoor $PM_{2.5}$ ($r = 0.70$, $P < 0.0001$). Personal measurements were generally higher than indoor measurements (median percentage difference 6.5%). Linear mixed effect models were constructed, using the same approach as detailed above in order to assess which variables contributed to the observed differences between the log of the personal and indoor measurements. They observed that colder temperatures

dx.doi.org/10.1021/es502201s | Environ. Sci. Technol. 2014, 48, 8456−8464

Table 2. Personal and Indoor $PM_{2.5}$ ($\mu g/m^3$) Exposure Related to Different Stove Ventilation Configurations and Fuel Types

		personal				indoor			
fuel type	stove design	N[a]	AM[b]	GM[b]	GSD[b]	N[a]	AM[b]	GM[b]	GSD[b]
smoky coal		206	180	148[c]	1.9	210	185	144[c]	2.0
	vented stove	110	150	134	1.6	114	149	127	1.7
	unvented stove[d]	8	252	233	1.6	8	221	183	2.0
	portable stove	22	178	143	1.9	20	168	135	2.0
	firepit	15	307	277[e]	1.6	15	371	350[e]	1.4
	mixed ventilation stove	44	219	164	2.3	45	232	166	2.1
smokeless coal		47	152	115	1.9	45	104	96[f]	1.6
	vented stove	5	151	126	2.0	5	117	104	1.7
	unvented stove	18	167	109	2.1	17	107	103	1.4
	portable stove	19	150	123	1.9	18	101	89	1.8
	firepit	3	104	102	1.3	3	91	90	1.3
	mixed ventilation stove	2	97	95	1.3	2	85	83	1.4
"mixed" coal[g]		38	183	161	1.7	42	164	130	2.0
	vented stove	13	152	137	1.7	14	151	123	1.9
	unvented stove	0				0			
	portable stove	14	209	180	1.8	14	173	121	2.4
	firepit	2	156	150	1.5	2	157	154	1.3
	mixed ventilation stove	9	192	176	1.6	12	170	145	1.8
wood		24	369	289[f]	2.1	24	393	327[f]	1.9
	vented stove	8	226	183	1.9	8	339	257	2.2
	unvented stove	0				0			
	portable stove	6	327	320	1.3	5	247	244	1.2
	firepit	10	508	392	2.4	10	520	467	1.7
	mixed ventilation stove	0				0			
plant materials[h]		13	284	225[c]	2.1	13	417	276[f]	2.6
	vented stove	3	123	109	1.8	3	80	76	1.4
	unvented stove	3	416	408	1.3	3	402	377	1.6
	portable stove	2	439	439	1.0	2	422	407	1.5
	firepit	4	146	138	1.5	4	617	382	3.0
	mixed ventilation stove	1	605	605	NA	1	658	658	NA
"mixed" fuel[I]		94	205	160[c]	2.0	113	210	152[c]	2.2
	vented stove	19	121	104	1.8	22	140	98	2.3
	unvented stove	17	306	250[e]	2.2	26	316	220[e]	2.5
	portable stove	7	219	203	1.5	7	204	196	1.3
	firepit	0				0			
	mixed ventilation stove	47	207	165	1.9	54	196	153	1.9

[a]Data for unknown ventilation stoves or unknown fuel type are not shown. [b]AM = Arithmetic Mean, GM = Geometric Mean, GSD = Geometric Standard Deviation. [c]Significant ($p < 0.05$) variation between stove ventilation designs within designated fuel type via ANOVA testing. [d]Refers to high and/or low stoves without any chimney. [e]$p < 0.05$ when compared with vented stove in same fuel type via Tukey HSD test. [f]$p < 0.05$ when compared with smoky coal via Tukey HSD test. [g]Refers to the use of combinations of smoky, smokeless coal, and prepared coal briquettes. [h]Plant materials include combinations of wood, tobacco stem, and corncob. [I]Refers to combinations of wood, plant materials and coal.

and the use of more fuel (as measured by weight) contributed to higher personal measurements, relative to indoor measurements (SI Table S2).

Descriptive statistics showed variations of $PM_{2.5}$ measurements between the various fuel types and stove designs (Table 2). In general, burning wood [GM(GSD):289 (2.1) and 327 (1.9) $\mu g/m^3$] or plant materials [GM(GSD):225 (2.1) and 276 (2.6) $\mu g/m^3$] resulted in the highest personal and indoor $PM_{2.5}$ levels, respectively. Significantly higher indoor $PM_{2.5}$ concentrations were observed among smoky coal burning homes compared to homes using smokeless coal [GM(GSD):144 (2.0) versus 96 (1.6) $\mu g/m^3$, $p < 0.05$]. Personal $PM_{2.5}$ exposure attributable to people using smoky coal was also higher than those using smokeless coal [GM(GSD):148(1.9) versus 115(1.9) $\mu g/m^3$], although the exposure did not differ significantly.

When assessing the role of stove design, we observed that measurements from homes and individuals using vented stoves were generally lower than if an unvented stove or firepit was used. Notably, among smoky coal burning homes, vented stoves had significantly lower personal and indoor $PM_{2.5}$ exposures as compared with firepits [personal values: GM(GSD):134 (1.6) versus 277 (1.6) $\mu g/m^3$, $p < 0.05$; indoor values: 127(1.7) versus 350(1.4) $\mu g/m^3$, $p < 0.05$]. Vented stoves were also observed to have significantly lower $PM_{2.5}$ levels than unvented stoves for homes burning "mixed" fuels [personal measurements: 104 (1.8) versus 250 (2.2) $\mu g/m^3$, $p < 0.05$; indoor measurements: 98(2.3) versus 220(2.5) $\mu g/m^3$, $p < 0.05$].

An assessment of variation in $PM_{2.5}$ levels between and within designated smoky coal subtypes is presented in Table 3. Only households that exclusively burned smoky coal were included in the analysis (199 personal and 210 indoor

Table 3. Personal and Indoor $PM_{2.5}$ ($\mu g/m^3$) Concentrations from Smoky Coal Burning Homes from Xuanwei and Fuyuan, by Coal Source

			personal				indoor			
county	smoky coal subtype	coal mine	N[a]	AM[b]	GM[b]	GSD[b]	N	AM	GM	GSD
Xuanwei	*coking coal*		*119*	*189*	*153*	*2.0*	*122*	*196*	*149*	*2.0*
		Azhi	34	227	181	1.9	33	186	160	1.7
		Baoshan	12	210	168	2.2	12	246	193	2.1
		Laibin	28	153	132	2.1	30	220	152	2.2
		Tangtang	31	194	152	2.0	33	191	133	2.3
		Yangchang	14	142	125	1.6	14	135	124	1.5
Fuyuan	overall		80	168	142[c]	1.8	88	169	138[c]	1.9
	coking coal		*23*	*213*	*175*[d]	*1.9*	*27*	*188*	*154*[d]	*1.9*
		Daping	9	111	104	1.5	10	87	83	1.4
		Enhong	9	241	208	1.8	11	250	222	1.7
		Haidan	5	348	329	1.4	6	242	221	1.7
	1/3 coking coal		*13*	*183*	*165*[d]	*1.6*	*12*	*271*	*245*	*1.7*
		Bagong	10	207	194	1.4	9	274	262	1.4
		Dahe	3	104	96	1.6	3	263	200	2.8
	gas fat coal		*40*	*135*	*120*[d]	*1.6*	*39*	*117*	*107*[d]	*1.6*
		Housuo	38	130	116	1.6	37	111	102	1.5
		Qingyun	2	237	237	1.0	2	236	235	1.1
	meager lean coal	Gumu	*4*	*138*	*96*	*2.8*	*4*	*196*	*149*	*2.0*

[a]Number of measurements is from households which exclusively burn smoky coal and report a coal source consistent with reported coal type. [b]AM = Arithmetic Mean, GM = Geometric Mean, GSD = Geometric Standard Deviation. [c]Significant ($p < 0.05$) variation between smoky coal subtypes sourced in Fuyuan via ANOVA test. [d]Significant ($p < 0.05$) variation between coal mines within identified smoky coal subtype via ANOVA test.

measurements). ANOVA analysis revealed little variation between the coking coal mines from Xuanwei but showed significant variation between the four designated smoky coal subtypes for both personal and indoor measurements (coking coal, 1/3 coking coal, gas fat coal, and meager lean coal) in Fuyuan. Furthermore, significant variation in personal $PM_{2.5}$ levels was observed for coal sourced from within the coking coal, 1/3 coking coal and gas fat coal mines in Fuyuan (indoor measurements showed significant variation in the coking and gas-fat coal subtypes).

Linear mixed effect modeling was carried out to identify variables that had a significant role in personal $PM_{2.5}$ exposures. Of the 32 variables considered, 5 were found to significantly impact personal $PM_{2.5}$. These determinants are the broad fuel types (smoky coal, smokeless coal, etc.), stove ventilation, the season of the year, the number of windows in the main cooking area, and the recorded number of hours burning a solid fuel standardized by the number of stoves used. Estimates (β), 95% confidence intervals, and geometric mean ratio [GMR = exp(β) = GM(estimate)/GM(reference)] are provided in Table 4.

The linear mixed effect model of personal $PM_{2.5}$ explains 35% of the variance between subjects and 79% of the variance between villages (Table 4). Among the assessed fuel types, wood results in the highest modeled $PM_{2.5}$ exposure (GMR: 2.80). Among stove designs, unvented stoves were found to result in the highest predicted $PM_{2.5}$ exposure (GMR: 1.62) although the use of firepits resulted in similar predicted $PM_{2.5}$ exposure (GMR: 1.47). Homes with one window in their main cooking area were found to result in higher $PM_{2.5}$ exposure (GMR for one window: 1.25) than those with zero or two windows (GMR for two windows: 0.99). The season during which measurements were taken also played a role in $PM_{2.5}$ exposure, with measurements taken in summer resulting in the lowest predicted $PM_{2.5}$ exposure (GMR: 0.71) and measurements taken in winter resulted in the highest (GMR: 1.21). The amount of time burning fuels, standardized by the number of stoves used, also played a role in $PM_{2.5}$ exposure with every hour of stove operation [median operating time 4.3 h; Interquartile Range (IQR) 2.2 to 9.6 h per stove] resulting in an incremental increase (GMR: 1.01) of $PM_{2.5}$ exposure. The model output for indoor measurements is very similar to personal measurement model, shown in SI Table S3.

DISCUSSION

The combustion of biomass and coals is a significant contributor to HAP and exposure to $PM_{2.5}$ worldwide. Biomass burning may account for 74–87% total $PM_{2.5}$ in households with a single dominant cooking source.[21] A wide range of factors, including stove design, fuel type, activity patterns, weather conditions, and household room configuration, can contribute to HAP.[20] For example, fuel type and ventilation were found to be significant determinants of household $PM_{2.5}$ in a study based in India,[22] while a Chinese based stove emission study showed that honeycomb coal resulted in lower emissions of sulfur dioxide, nitrogen oxide, and total suspended particulate, but 2–3 fold higher $PM_{2.5}$, compared to coal cake.[23] In our study on personal and indoor $PM_{2.5}$ exposure in a rural population in China with high lung cancer incidence, fuel type, ventilation, cooking room configuration, season, and burning time per stove were identified as main determinants of $PM_{2.5}$ exposure.

Our study found that burning wood and other plant materials in unvented stoves resulted in the highest $PM_{2.5}$ measured both personally and indoors. These measurements are consistent with international research in similar settings for unvented stoves using wood [albeit our measurements are at the lower end of the exposure range (AM for indoor $PM_{2.5}$ air measurements 520 $\mu g/m^3$)]. For example, the AM of 22-h $PM_{2.5}$ measurements of 9 households in kitchens with an open wood stove in Guatemala was 528 ± 249 $\mu g/m^3$.[24] The geometric mean 48-h $PM_{2.5}$ of 53 households in kitchens using wood in an open fire without ventilation in rural Mexico was

Table 4. Linear Mixed Effect Modeling of ln-Transformed Personal $PM_{2.5}$ Exposure

	estimate (β)	95% CI	GMR[a]
fuel type			
smokeless coal	ref.		1.00
smoky coal	0.27	0.02,0.52	1.31
"mixed" coal	0.35	0.06,0.64	1.42
wood	1.03	0.66,1.40	2.80
plant materials	0.43	0.02,0.84	1.54
"mixed" fuel	0.37	0.11,0.63	1.45
stove design			
vented stove	ref.		1.00
unvented stove	0.48	0.22,0.74	1.62
portable stove	0.26	0.06,0.47	1.30
firepit	0.38	0.10,0.66	1.47
mixed ventilation stove	0.2	0.03,0.36	1.22
unknown ventilation stove	−0.34	−0.77,0.09	0.71
number of windows in main cooking room			
none	ref.		1.00
one	0.22	0.01,0.44	1.25
two	−0.01	−0.26,0.23	0.99
season			
autumn	ref.		1.00
winter	0.19	0.02,0.36	1.21
spring	−0.24	−0.41,-0.07	0.79
summer	−0.34	−0.68,0.00	0.71
number of hours burning fuel standardized by number of used stoves[b]	0.01	0.003,0.03	1.01
variation explained, %			
between individual subjects		35	
between villages		79	
reference value[c], ln-μg/m^3		4.35	

[a]Geometric mean ratio = GM(estimate)/GM(reference) = Exp(β). [b]Median period 4.3 h; IQR 2.2 to 9.6 h per stove. [c]Reference value represents base value of log transformed $PM_{2.5}$ in model for reference group (smokeless coal burnt in a vented stove, during autumn in a room with no windows).

615 μg/m^3,[25] and the AM of 48-h $PM_{2.5}$ measurements of 63 households in the highlands of San Marcos, Guatemala using open wood fires was 900 ± 700 μg/m^3.[26]

$PM_{2.5}$ exposure levels among coal users in our study were similar to other studies studying coal combustion for cooking and heating in China. An indoor air pollution measurement study in four provinces in China observed indoor Respirable Particulate Matter (RPM) concentrations in the primary biomass fuel provinces of Inner Mongolia and Gansu of 719 and 661 μg/m^3, respectively, while indoor RPM concentrations in the primarily coal-burning provinces of Guizhou (202−352 μg/m^3) and Shanxi (187−361 μg/m^3) were lower.[27] The Sino-Dutch project including 150 households in five counties of three provinces monitored 24-h $PM_{2.5}$ levels averaging 290 μg/m^3 in households mainly burning coal.[28]

The observed differences between studies could be caused by several factors including differences in coal types. In our study, we observed differences in $PM_{2.5}$ emissions between smoky and smokeless coal (GM for indoor measurements: 144 μg/m^3 and 96 μg/m^3 respectively). Furthermore, when investigating for variation in $PM_{2.5}$ measurements between smoky coal sources, we observed significant variation between smoky coal subtypes from Fuyuan. Variation was also observed between coal mines producing the same smoky coal subtypes for the coking, 1/3 coking and gas fat coal subtypes. This indicates that when comparing HAP exposure studies the exact fuel types need to be carefully considered. It also provides indications that the observed health effects may be differential within a particular solid fuel type (e.g., coals).[29]

Many of the previous exposure studies on HAP have focused on measuring indoor air concentrations with relatively few measuring personal exposure, resulting in uncertainty in how these indoor measurements correspond to personal exposure.[20] Several studies have shown that indoor air measurements do not always accurately reflect personal exposure levels to HAP, possibly limiting the interpretation of these indoor measurements.[30,31] In our study, we found that personal and indoor $PM_{2.5}$ air measurements were quite similar and that the correlation coefficient between indoor air $PM_{2.5}$ concentrations and personal $PM_{2.5}$ exposure levels was high (Spearman r = 0.70, $P < 0.0001$). This association was modified primarily by the volume of fuel used and temperature (i.e., season) (SI Table S2), both likely proxies of residence time inside the home. This suggests that indoor air concentrations may be good proxies for personal exposure levels of nonsmoking females in this study area. The generalizability of this observation may be limited as our enrollees were generally older women (mean age of 56 years) who were primarily responsible for cooking and housekeeping, meaning that they generally spent the majority of their days indoors. Men, and younger women, who may spend a greater portion of their time outdoors may have different exposure experiences and a weaker correlation between indoor and personal $PM_{2.5}$ exposure levels.

Mixed effect modeling showed that fuel type, ventilation, number of windows, and season are strong determinants of personal $PM_{2.5}$ exposure, which reflects some of the findings of the descriptive statistics (specifically fuel types and stove design). Ventilation has been shown to be effective in reducing HAP exposures[24,25,30,32]and in reducing malignant and non-malignant disease both internationally and in Xuanwei and Fuyuan. We observed a difference of 34−80% in $PM_{2.5}$ concentrations between vented stoves and unvented stoves/firepits burning smoky coal, wood, plant materials and combinations of coal/plant materials. These values are similar to the reduction reported by the Sino-Dutch project in China where a reduction of 40% was observed after implementation of stove improvements,[33] and slightly lower than what has been reported for stove improvement programs in Latin America where reduction levels ranging between 70% and 80% have been reported among primarily wood burning homes.[24,25,30,32] Additionally, although our study is using a cross-sectional design, our households were selected based on having no stove improvements in the last 5 years. As such the role of stove ventilation presented here represents designs that have been continually used for many years and have likely undergone some "wear and tear". Therefore, this may result in somewhat lower decreases than observed in other studies that performed their evaluations shortly after the introduction of stove improvements. However, this also shows that stove improvement programs do not only result in a short-term reduction in HAP but one which seems to be sustained for a longer time. The finding of increased personal $PM_{2.5}$ in the colder seasons is consistent with expected behavior, as people would spend a greater proportion of their time indoors.

The finding that having one window in the main cooking room presented higher modeled $PM_{2.5}$ measurements than those with zero or two windows seems counterintuitive to the logical expectation. A possible explanation for this observation may relate to home design. If we postulate based on field observations that homes with zero windows represent poorer households, then it is likely that they have poorer construction than homes with windows. This poorer construction may increase home ventilation due to imperfections in the structure of the home acting as ventilation conduits. The role of burning time, standardized by the number of stoves used in personal exposure to $PM_{2.5}$ is consistent with the expectation of increased exposure with increased burning time.

The findings of this work provide a valuable insight into potential etiological factors of the lung cancer epidemic in Xuanwei and Fuyuan. However, the high $PM_{2.5}$ measurements in emissions from wood and plant burning homes (which do not have high lung cancer rates when compared to smoky coal burning homes) indicate that measurements of $PM_{2.5}$ exclusively will not be sufficient to explain the lung cancer epidemic in Xuanwei and Fuyuan. Future research will further investigate the constituents of fuel emissions and work toward associating those emissions with lung cancer epidemiology.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Table S1, Variables considered for inclusion in mixed model creation. Table S2, Linear mixed effect model indicating variables which contribute to differences between indoor and personal measurements. Table S3, Linear mixed effect modelling of ln-transformed indoor PM2.5 exposure. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This project was supported by the National Institutes of Health intramural research program. The content of this publication does not necessarily reflect the views or policies of the Department of Health and Human Services, nor does mention of trade names, commercial products, or organizations imply endorsement by the U.S. Government. The authors would like to acknowledge Jackie King from BioReliance and Peter Hui and Nathan Appel from Information Management Systems, Inc. for their support.

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