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Polycyclic Aromatic Hydrocarbon Exposure in Household Air Pollution from Solid Fuel Combustion among the Female Population of Xuanwei and Fuyuan Counties, China

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Supporting Information

ABSTRACT: Exposure to polycyclic aromatic hydrocarbons (PAHs) from burning "smoky" (bituminous) coal has been implicated as a cause of the high lung cancer incidence in the counties of Xuanwei and Fuyuan, China. Little is known about variations in PAH exposure from throughout the region nor how fuel source and stove design affects exposure. Indoor and personal PAH exposure resulting from solid fuel combustion in Xuanwei and Fuyuan was investigated using repeated 24 h particle bound and gas-phase PAH measurements, which were collected from 163 female residents of Xuanwei and Fuyuan. 549 particle bound (283 indoor and 266 personal) and 193 gas phase (all personal) PAH measurements were collected. Mixed effect models indicated that PAH exposure was up to 6 times higher when burning smoky coal than smokeless coal and varied by up to a factor of 3 between different smoky coal geographic sources. PAH measurements from unventilated firepits were up to 5 times that of ventilated stoves. Exposure also varied between different room sizes and season of measurement. These findings indicate that PAH exposure is modulated by a variety of factors, including fuel type, coal source, and stove design. These findings may provide valuable insight into potential causes of lung cancer in the area.

INTRODUCTION

Xuanwei and its neighboring county of Fuyuan, located in Yunnan province, China, have among the nation's highest lung cancer rates in both men and women, irrespective of smoking status.[1−3] Previous research has associated the domestic combustion of locally sourced "smoky" (bituminous) coal with this excess cancer rate.[4,5] Solid fuels are used for heating and cooking throughout Xuanwei and Fuyuan, of which coal is the most common (alternative fuels include wood, corn cobs, and tobacco stems). There are multiple active coal mines throughout both counties with mines typically producing either smoky or "smokeless" (anthracite) coal (the terms smoky and smokeless refer to the amount of visible smoke released on combustion). Historically, residents have typically purchased coal from their nearest mine.

Two major features have been observed to drive lung cancer rates among those burning smoky coal. The first feature relates to stove design. Historically people burnt fuel in unvented firepits. In recent decades, these have been replaced with a variety of differing stove designs which have the purpose of more efficient burning characteristics and reducing household air pollution (HAP). These improved stoves have resulted in reduced cancer rates[6,7] (as well as reduced nonmalignant lung disease),[8] indicating that exposure to carcinogenic material(s) is reduced through these designs. The second feature is the observation that lung cancer rates among smoky coal users vary (by up to 20 times) between geographic locations.[4,9] Given that lifestyle factors are largely similar throughout Xuanwei and Fuyuan, this suggests that there may be constitutional differences between coals sourced from different locations, whether in the geological formation, or in varying practises of coal preparation (e.g., briquetting). Evidence to support geological differences in coal from different areas comes from

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the State Standard of China Coal Classification which recognizes at least four "sub-types" of smoky coal in the region.[10] These subtypes are based upon a variety of criteria including the degree of coalification (measured as the dry ash free volatile matter) and the caking property of the coal (which is a combination of the caking index, the maximum thickness of the plastic layer and the Audibert-Arnu dilation).[10,11]

Research investigating the properties of uncombusted coal, collected from coal mines and from homes of Xuanwei and Fuyuan residents has shown that smoky coal contains high amounts of volatile organic compounds (i.e., hydrocarbon content) and quartz but generally low levels of trace elements when compared to smokeless coal.[11] Research investigating the emissions of coal combustion has found that smoky coal (compared to smokeless coal) emits high amounts of nanoparticles,[12] particulate matter,[13] and polycyclic aromatic hydrocarbons (PAHs), specifically benzo[a]pyrene (BaP).[1] BaP is considered a known carcinogen by the International Agency for Research on Cancer (IARC).[14] Stove improvements have resulted in reduced indoor BaP levels, which parallels observations of reduced lung cancer rates following stove improvement. However, reduced BaP exposure has not been explicitly linked to reduced cancer rates and ventilation has also been shown to reduce other components than BaP.[7,15,16] Additionally, variation in indoor BaP levels has been observed between different geographic locations in Xuanwei,[2] reflecting to some extent the geographic variability in lung cancer rates. However, the geographic variations in BaP exposure observed were related to a variety of fuel sources and stove designs and have not explicitly been linked to a single fuel type. Furthermore, the bulk of the published research thus far has been limited by small sample sizes, limited geographical scope, limited number of PAHs assessed and a reliance upon the use of HAP measurements as a proxy for personal exposure.

The goal of this paper is to investigate indoor and personal exposure to PAHs resulting from domestic solid fuel combustion in Xuanwei and Fuyuan homes. Particular attention will be paid to the relationship between different fuel types, fuel sources, and different stove designs.

■ MATERIALS AND METHODS

Study Design. This paper is part of a cross-sectional epidemiology study aimed at comprehensively cataloguing the constituents of smoky coal and other solid fuels used in Xuanwei and Fuyuan and associating those constituents with lung cancer risk and early biological effect markers in a case-control study of lung cancer among never-smoking females. The full details are provided elsewhere[11] but briefly: 15 villages were selected from each county. Up to five houses were selected from each village and a nonsmoking female between the ages of 20 and 80 from each house was enrolled for personal monitoring of airborne pollutants and activity monitoring. All study participants provided written informed consent prior to their enrolment in the study. At all stages, selection was targeted to represent the population present in the case-control study with regard to age and typical living arrangements and therefore would also reflect historical stove and fuel usage in the area, thus houses were preferentially selected for enrolment if they were at least ten years old and had not altered their stoves for at least the past five years. Each house was measured and sketched. Stoves and other pertinent features (e.g., doors, windows and stairways) were recorded. Study enrolees reported current and historical fuel and stove usage in addition to medical histories and social demographic information.

Data was collected over two time periods, August 2008 to February 2009 and March to June 2009. In the first collection period, all 30 villages were visited, with 148 participants recruited. In the second, 16 villages were revisited (villages selected reflected the overall population) with 53 of the initial subjects resampled and 15 new subjects recruited. During each period, samples were taken during two consecutive 24 h periods.

Stove and Fuel Information. During each sampling period, subjects activities were documented. When a subject used a particular coal type (smoky or smokeless), geochemical analysis on the solid coal[11] was used to confirm the coal classification. Subjects using coal reported their supplying coal mine. Smoky coal subtypes were identified by linking the Chinese State Standard coal classification to the reported mine.

Following a review of the activity logs, descriptive categories for fuel and stove design were established. For fuel usage, the categories were: smoky, smokeless, "other" coal (referring to combinations of coal types and usage of processed coal products such as briquettes), wood, plant products (referring to combinations bamboo shoots, tobacco stems and corn cobs, sometimes in combination with wood) and "other" fuel (referring to combinations of coal and plants). Only homes which exclusively used smoky or smokeless coal were classified as such.

Stove categories used for final analysis are ventilated stoves, unventilated stoves, firepits, portable stoves (stoves designed to be lit outside and then carried indoors for use), "mixed" (usage of multiple types of stoves with differing ventilation) and "unknown" (where stove type was not recorded).

Sample Collection and Analysis. The PAHs assessed were a combination of particle bound (high molecular weight) and gas phase (low molecular weight) PAHs. The particle bound PAHs analyzed were: fluoranthene (FLT), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]flouranthene (BkF), benzo[a]pyrene (BaP), dibenz(ah)anthracene (DBA), benzo[ghi]perylene (BPE), and indeno(1,2,3-cd)pyrene (IPY). The gas phase PAHs assessed were: fluorine (FLU), naphthalene (NAP), acenaphthylene (ANY), phenanthrene (PHE), acenaphthene (ANA), and anthracene (ANT).

Particle bound PAHs were measured through the use of personal and indoor measurements. Particulate matter was collected on a 37 mm Teflon filter using a cyclone with an aerodynamic cutoff of 2.5 μm, powered by a BGI AFC400S pump. Pumps were calibrated prior to each measurement. Flow rates were measured pre and postmeasurement and samples were not accepted if the flow rates varied by more than 10%.The median flow rate was 3.3L/min (interquartile range: 3.24−3.47L/min). For personal measurements, the pumps were carried by participants with the cyclone attached in proximity to their breathing zone. Overnight, personal devices were placed adjacent to subjects beds. For indoor measurements, pumps were placed approximately 0.25 m from the wall and between 1 and 2 m from the main stove (as allowed by the size of the room).

Personal gas phase PAHs were measured through the use of XAD-2 sorbent tubes at a median air flow rate of 63 mL/min (interquartile range: 47−80 mL/min).

Filters and XAD-2 resin were inserted in separate extraction tubes after which 5 mL of dichloromethane (DCM) was added

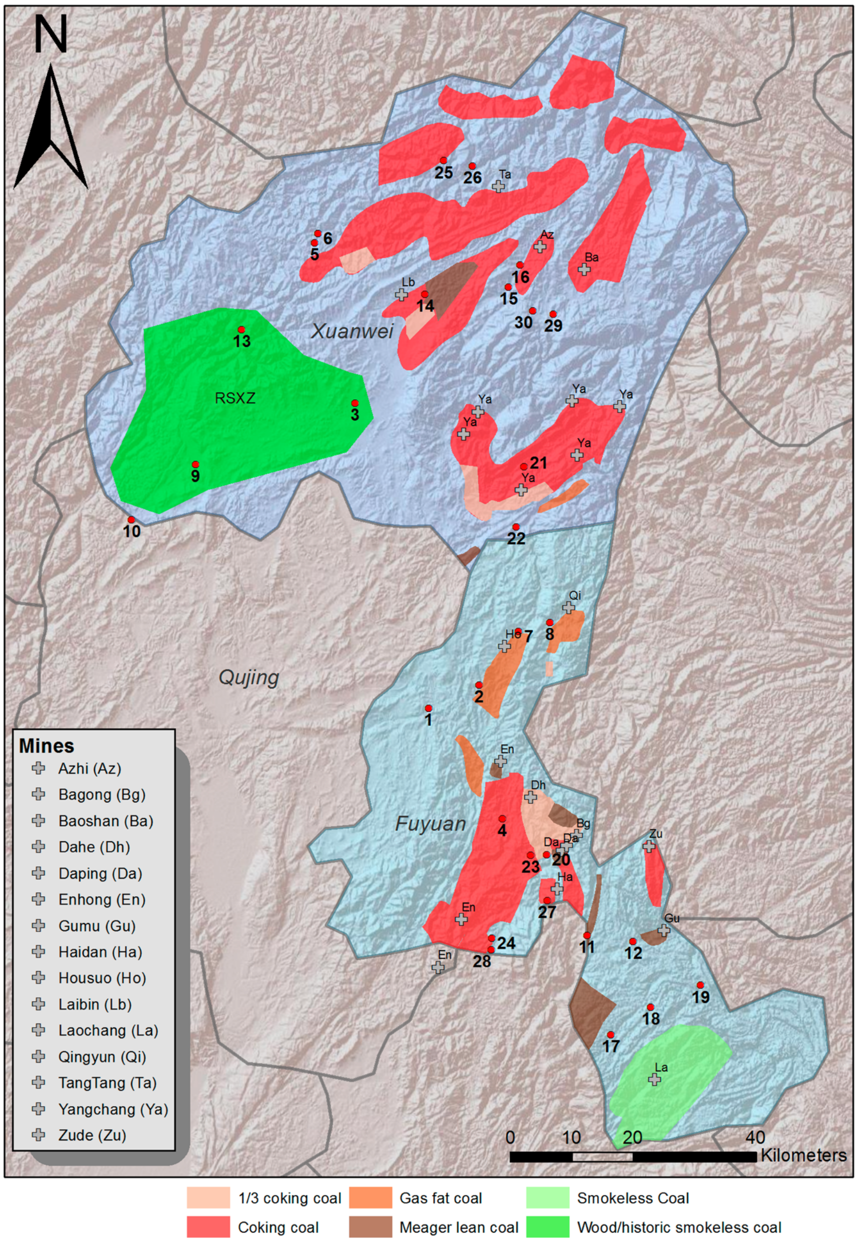

Figure 1. Map of Xuanwei and Fuyuan counties. Village location is indicated by designated numbers. Mines indicated are those reported by study participants and do not represent all mines present in the area.

and the tube was capped. Tubes were ultrasonically extracted for 60 min at 60°C. After cooling to room temperature, the extract was filtered into a sampling vial using a 0.45 μm polytetrafluoroethylene (PTFE) membrane filter. Extraction recoveries were determined by spiking the pre-extracted sample with 500 nL (100 μL of 5 ng/ μL) of six deuterated internal standards containing Acenaphthene-d10, Chrysene-d12 (CHR-D12), 1,4-dichlorobenzene-d4 (1,4-DCB-D4), naphthalene-d8, perylene-d12, and phenanthrene-d10.

A gas chromatograph connected to a mass spectrometer (Shimadzu QP2010 plus) was used to determine the 16 PAH species. Separation of the compounds was carried out on a DB-5MS columns (Agilent 30 m × 0.32 mm × 0.25 μm). Target PAHs were identified based on the retention time and qualitative ions of the standards in a selected ion monitoring mode that were quantified by the internal standards. The quantity of each PAH was calculated in nanograms. The limit of detection was set to 12.5 ng (0.00025 ug/mL). Only values for gas-phase PAHs collected from XAD-2 resin and particle bound PAHs collected from Teflon filters were retained for statistical analysis (the median nondetect rate for particle bound PAHs on XAD-2 resin and gas phase PAHs on Teflon filters was 97%).

For quality control purposes, field blank samples were analyzed in conjunction with the exposed filters. Field blanks reported nondetect measurements for greater than 97% of measurements, thus no blank correction was required. The average recovery rate of internal standards was 96%. Median recovery rates ranged from 87% (1,4-DCB-D4) to 101%(CHR-D12). In view of the high recovery we did not correct results for recovery efficiency. We analyzed 13 field duplicate samples for particle and gas bound PAH's. The coefficient of variation for duplicate PAH measurements ranged from 7% (CHR) to 47% (PHE) with a median value of 25%.Values for total PAH

Table 1. Exploratory Factor Analysis of Log Transformed Personal PAH Values[a]

	particle phase			gas phase	
	Factor 1	Factor 2	Factor 3		Factor 1
EV(% variance explained)	4.5 (45%)	3.7 (37%)	1.3 (13%)	EV(% variance explained)	3.7 (91%)
benzo[ghi]perylene (6)	**0.81**	0.44	0.29	naphthalene(2)	**0.94**
benzo[b]fluoranthene (5)	**0.70**	0.47	**0.51**	phenanthrene (3)	**0.98**
dibenz[ah]anthracene(5)	**0.80**	0.29	0.30	acenaphthylene (3)	**0.90**
benzo[a]pyrene (5)	**0.73**	**0.60**	0.30	fluorine (4)	**0.99**
benzo[k]fluoranthene (5)	**0.72**	**0.56**	0.33		
indeno[1,2,3-cd]pyrene (5)	**0.83**	0.48	0.18		
pyrene(4)	0.43	**0.87**	0.25		
fluoranthene(4)	0.39	**0.88**	0.25		
chrysene(4)	**0.56**	**0.57**	**0.60**		
benzo[a]anthracene (4)	**0.62**	**0.65**	0.41		

[a]Values in bold (Eigen value >0.5) are considered to be contributory to that factor. Numbers in parentheses represent number of carbon rings for respective PAH. Factor analysis was performed with varimax rotation.

content (sum of PAHs) and BaP equivalent (calculated using previously reported toxic equivalency factors for PAHs[17]) were calculated and are available in the supplement.

Statistical Analysis. Levels of ANT and ANA were below the limit of detection for 78% of the samples collected and were excluded from statistical analysis. Of the remaining PAHs, between 78% (FLT) and 98% (BbF) of the particle bound PAHs and between 68% (FLU) and 95% (NAP) of the gas phase PAHs were above the limit of detection (median detection rates of 93% and 80% for particle-bound and gas phase measurements respectively). Measurements below the limit of detection were imputed from a log-normal probability distribution via a multiple imputation procedure.[18] PAH concentrations were calculated by dividing the measured PAH value (in ng) by the total volume of air (in m^3) drawn through either filters or the XAD2 tubes.

Normal probability plots indicated that PAH values followed a log-normal distribution. Descriptive statistics included arithmetic means (AM) geometric means (GM) and geometric standard deviations (GSD). ANOVA and Tukey HSD testing on log-transformed PAH values was performed to assess for variation between fuel types and stove types.

The presence of possible latent exposure variables was explored with factor analysis on log-transformed values using varimax rotation to provide noncorrelated factors.[19] Factors with an eigen value greater than one were retained and individual variables within each identified factor with loading values of greater than 0.5 were considered to significantly contribute to that factor. Subsequently, a single PAH from each factor was selected to act as a proxy for its respective factor in the construction of predictive linear mixed effect models. Model construction was targeted toward a single, parsimonious model which could be applied to each proxy PAH. Villages and individual subjects were assigned as random effects. Variables considered for inclusion as fixed effects included fuel type, fuel source, weight of fuel used, stove design, room size, number of windows/doors, the presence of stairways, meteorological conditions, and the season when measurements were taken. A list of all considered variables is available in Supporting Information (SI) Table S1). The following formula can be used to summarize the model:

$$y_{iif} = \mu + \beta_1 x_1 + \beta_2 x_2 ... \beta_n x_n + bI_i + bJ_{ij} + \varepsilon_{iif}$$

Where y_{ijf} represents the natural log transformed value of the PAH exposure levels for village i, person j on day f. μ represents the intercept (i.e., the "background" level). β_1 through β_n represent fixed effect variable coefficients for variables x_1 through x_n. b_{Ii} represents the random effect coefficient for village i. b_{Jij} represents the random effect coefficient for subject j, living in village i. ε_{ijf} represents the error for village i, person j on day f.

All statistical testing was carried out in R version 3.03[20] using the lme 4 package.[21] p values less than 0.05 were considered to indicate statistical significance.

RESULTS

Overview. In total, 163 subjects were recruited. 549 measurements of particle phase PAHs (283 indoor and 266 personal measurements) were collected. 268 of these (137 indoor and 131 personal) reflected exclusive use of smoky coal. Of that 268, 258 (132 indoor and 126 personal) could be associated with an individual coal mine. 193 personal measurements of gas phase PAHs were taken, of which 96 reflected smoky coal use, 93 of which could be associated with an individual coal mine. A map of Xuanwei and Fuyuan, containing the locations of enrolled villages, reported coal mines and coal subtypes is shown in Figure 1. In Xuanwei, all smoky coal used was the subtype "coking coal". Smoky coal from Fuyuan consisted of the "coking coal", "$^1/_3$ coking coal", "meagre lean coal", and the "gas fat coal" subtypes.[10,11]

Indoor and personal particle phase PAH measurements showed a moderate to good correlation, with Spearman correlation coefficients ranging from 0.56 (DBA) to 0.80 (BaA) with a median correlation value of 0.76 (Pearson correlation of log-transformed values: 0.56 [DBA] to 0.82 [FLT], median 0.76). Results of personal measurements were generally slightly lower than indoor measurements (2%). However, we observed the difference between personal and indoor measurements was influenced by the season of measurement. During winter, the median difference between personal and indoor measurements ranged from personal being 19% higher (BaA) to 8% lower (DBA) with an overall median difference of personal measurements being 7% higher than indoor. In contrast, during the spring and summer months, personal measurements were on median 8% lower than indoor measurements.

Factor Analysis. The results of factor analysis are shown in Table 1. Particle bound and gas phase personal PAH

Table 2. Personal Exposure to Selected PAHs (in ng/m^3) by Fuel Type and Stove Design[a]

		BaP			FLT			CHR				NAP		
	N(k)	AM	GM	GSD	AM	GM	GSD	AM	GM	GSD	N(k)	AM	GM	GSD
smoky coal	**131(82)**	**74.4**	**44.7**	**2.8**	**65.3**	**19.3**	**4.7**	**94.3**	**44**	**3.6**	**96(60)**	**4200**	**2900**	**2.4**
ventilated stove	72	50.2	38.1	2.1	28.7	15.6	3.3	54.3	34.5	2.8	53	2900	2500	1.9
unventilated stove	6	224.5	160.3	2.4	273.6	108.2	5.1	374.8	217.3	3.2	4	11 000	6300	3
portable stove	13	41.5	31.5	2.2	17.2	9.2	3.2	68.7	45	2.7	12	4900	3900	2.1
firepit	11	186.4	151.5[c]	2	228.7	147.6[c]	2.7	245.2	186.4[c]	2.1	4	17 000	14 000	2.1
mixed ventilation	25	85.7	48.1	3.2	84.1	21.9	5.4	103	45.5	4.1	19	3900	2600	3.2
unknown	4	13.2	7.7	3	1.6	1.4	1.8	7.9	4.4	3.2	4	1300	1100	2
smokeless coal	**27(16)**	**15.1**	**10.6[b]**	**2.5**	**5.6**	**2.8[b]**	**3.2**	**29.3**	**12.6[b]**	**3.5**	**17(11)**	**3100**	**2800**	**1.6**
ventilated stove	2	5.6	5.5	1.3	3.9	2.7	3.7	7	4.8	3.8	0			
unventilated stove	14	13.8	9.4[b]	2.7	2.4	1.7[b]	2.2	12.6	8.3[b]	2.6	9	2800	2600	1.6
portable stove	10	19.3	14.2	2.4	10.7	5.4	3.9	58.7	27.3	4	7	3600	3400	1.5
firepit	0										0			
mixed ventilation	1	10.6	10.6		4.2	4.2		13.7	13.7		1	2000	2000	
unknown	0										0			
"other" coal	**24(16)**	**61.5**	**39.7**	**2.8**	**44**	**15.6**	**5.5**	**89.1**	**43.2**	**3.9**	**14(10)**	**8300**	**5400**	**2.8**
ventilated stove	10	38.8	26.9	2.6	23.7	8.4	4.8	39.1	21.9	3.4	6	3200	2500	2.3
unventilated stove	0										0			
portable stove	9	95.6	68	2.8	65.1	31.1	5.2	159.4	91.1	3.8	7	13 000	11 000	2.1
firepit	1	21.8	21.8		3.3	3.3		7.6	7.6		0			
mixed ventilation	4	51.7	36.4	3.2	57.4	23.7	7.6	76.2	67.4	1.8	1	5900	5900	
unknown	0										0			
wood	**14(8)**	**66.6**	**58.2**	**1.7**	**73.7**	**34.7**	**4**	**66.2**	**54.3**	**2**	**7(4)**	**14 000**	**5900**	**6.4**
ventilated stove	5	73.4	61.2	1.9	73.5	19.1	6.2	73.3	51.6	2.9	2	13 000	13 000	1.3
unventilated stove	0										0			
portable stove	4	78.4	70.1	1.7	41.2	33.5	2.4	72.2	60	2	3	21 000	14 000	3.1
firepit	5	50.2	47.7	1.4	99.8	64.8	3.2	54.4	52.7	1.3	2	2000	730	10.9
mixed ventilation	0										0			
unknown	0										0			
plant	**4(4)**	**95.6**	**83.8**	**1.9**	**144.8**	**118.5**	**2.2**	**128.7**	**121.5**	**1.5**	**3(3)**	**26 000**	**20 000[b]**	**2.4**
ventilated stove	0										0			
unventilated stove	1	116.1	116.1		170	170		171.2	171.2		1	54 000	54 000	
portable stove	1	39.5	39.5		104.7	104.7		91.9	91.9		0			
firepit	1	67.3	67.3		42.2	42.2		81	81		1	10 000	10 000	
mixed ventilation	1	159.5	159.5		262.1	262.1		170.9	170.9		1	15 000	15 000	
unknown	0										0			
"other" fuel	**66(35)**	**73.8**	**36.9**	**3.3**	**82.6**	**17.4**	**6.6**	**96.3**	**37.3**	**4.3**	**56(28)**	**7600**	**4300**	**3.1**
ventilated stove	12	37.5	26.5	2.6	32.4	8.6	5.5	33.6	17.8	3.5	9	4500	3100	2.4
unventilated stove	13	158.8	73	4.7	197.2	42.9	10.7	242.5	102.3	5.5	11	20 000	13 000	3.4
portable stove	6	62	40.9	2.9	37.8	17.6	4.9	81.6	49.1	3.2	5	4400	4000	1.6
firepit	0										0			
mixed ventilation	32	60	33.5	3	70	17.4	5.6	70.8	35.5	3.5	28	4500	3300	2.7
unknown	3	21.3	16.5	2.5	12	5.4	5.4	15.7	8.7	4.2	3	4400	2500	3.7

[a]Mixed ventilation, use of multiple stoves with differing ventilation designs. *N*, number of samples. *k*, number of individual subjects. BaP, benzo[a]pyrene; FLT, fluoranthene; CHR, chrysene; NAP, naphthalene. [b]Significant difference with smoky coal for same PAH and strata (Tukey HSD test). [c]Significant difference with ventilated stove for same PAH within fuel strata (Tukey HSD test).

measurements were analyzed separately. Three factors were identified among the particle bound PAHs while all four of the gas phase PAHs contributed to a single factor. Among the factors identified from the particle phase PAHs, the first factor consisted primarily of PAHs with 5 and 6 rings: BPE, BbF, DBA, BaP, BkF, IPY, CHR, and BaA. The second factor consisted primarily out of PAHs with 4 and 5 rings: BaP, BkF, PYR, FLT, CHR, and BaA. The third factor consisted of CHR and BbF (4 and 5 rings, respectively)

Descriptive Statistics. One PAH from each factor described above was selected as a proxy for that factor and for the displaying of descriptive statistics. Descriptive statistics for the complete data set is available in the supplement. BaP was selected to represent the first factor. This selection was based upon the relative high loading of BaP (0.73), previous

Table 3. Determinants of Personal BaP, FLT, CHR, NAP Exposure[a]

	BaP			FLT			CHR			NAP		
reference/background value (intercept)[b]		2.55			1.45			2.4			7.45	
	estimate	95% CI	GMR	estimate	95%CI	GMR	estimate	95%CI	GMR	estimate	95%CI	GMR
Fuel Type												
smokeless coal (FY and XW)	ref		1	ref		1	ref		1	ref		1
coking coal from north XW	1.1	0.46,1.73	3.01	1.02	0.08,1.95	2.77	1.34	0.59,2.08	3.83	0.12	−0.53,0.76	1.12
coking coal from south XW	0.57	−0.53,1.66	1.77	0.47	−1.19,2.1	1.6	0.59	−0.65,1.84	1.8			
coking coal from FY	0.9	0.13,1.67	2.47	0.71	−0.42,1.84	2.04	1.21	0.29,2.12	3.35	−0.16	−0.53,0.76	0.85
1/3 coking coal FY	0.23	−0.72,1.14	1.26	0.48	−0.94,1.85	1.61	0.76	−0.34,1.81	2.14	−0.03	−1.16,0.86	0.97
gas fat coal FY	0.26	−0.51,1.01	1.3	0.19	−0.94,1.3	1.21	0.12	−0.76,0.98	1.13	−0.06	−1.34,1.28	0.94
meagre lean coal FY	0.82	−0.57,2.2	2.26	0.16	−1.88,2.19	1.18	1.15	−0.5,2.79	3.15	0.66	−0.86,0.71	1.93
multiple coal types	0.86	0.27,1.44	2.35	0.86	−0.01,1.73	2.37	1.07	0.38,1.75	2.9	0.59	−0.64,1.95	1.8
multiple fuel types	0.81	0.25,1.36	2.24	0.95	0.12,1.77	2.57	1.07	0.42,1.72	2.92	0.58	−0.11,1.28	1.78
smoky coal of uncertain source[c]	1.23	0.32,2.15	3.42	1.68	0.34,3.03	5.38	1.82	0.72,2.92	6.16	0.53	−0.05,1.19	1.7
plant products	0.03	−1.13,1.19	1.03	−0.22	−1.94,1.52	0.8	0.5	−0.89,1.9	1.65	0.45	−0.32,1.39	1.57
wood	0.44	−0.51,1.39	1.55	0.3	−1.08,1.7	1.35	0.9	−0.23,2.05	2.47	−0.55	−0.95,1.86	0.58
Stove Design												
ventilated stove	ref		1	ref		1	ref		1	ref		1
unventilated stove	0.43	−0.04,0.9	1.54	0.34	−0.36,1.04	1.41	0.62	0.05,1.19	1.86	0.72	−1.76,0.66	2.06
fire pit	1.08	0.4,1.76	2.94	0.16	0.73,2.71	5.62	1.06	0.24,1.87	2.9	1.37	0.1,1.02	3.95
portable stove	0.18	−0.24,0.6	1.2	1.73	−0.46,0.78	1.18	0.66	0.15,1.16	1.93	0.56	0.16,1.28	1.76
mixed ventilation	0.02	−0.3,0.35	1.02	0.11	−0.37,0.59	1.12	0.17	−0.23,0.56	1.18	−0.08	0.49,2.25	0.93
unknown ventilation	−1.01	−1.75,-0.25	0.37	−1.33	−2.43,-0.23	0.26	−1.27	−2.17,-0.37	0.28	−0.58	−0.44,0.29	0.56
Room Size (in m^3)												
<40 m3	ref		1	ref		1	ref		1	ref		1
40 m^3 to 49 m^3	−0.19	−0.61,0.24	0.83	0.62	−0.47,0.75	1.15	−0.46	−0.97,0.05	0.63	−0.2	−0.19,0.42	0.82
50 m^3 to 67 m^3	0.56	0.1,1.01	1.74	−0.63	0.21,1.54	2.39	0.49	−0.06,1.04	1.63	0.49	−0.68,0.27	1.64
>67 m^3	0.18	−0.28,0.64	1.2	0.14	−0.2,1.13	1.59	−0.04	−0.6,0.51	0.96	0.13	−0.02,0.97	1.14
unknown	0.12	−0.31,0.56	1.13	0.87	−0.28,0.99	1.42	−0.09	−0.61,0.44	0.92	0.05	−0.36,0.61	1.06
Season												
autumn	ref		1	ref		1	ref		1	ref		1
winter	0.17	−0.14,0.48	1.19	0.46	0.15,1.09	1.86	0.41	0.04,0.77	1.51	0.53	−1.27,0.12	1.7
spring/summer	−0.06	−0.33,0.21	0.94	0.35	−1.04,-0.23	0.53	−0.28	−0.6,0.03	0.75	0.11	0.14,0.9	1.12
Variance Explained												
between subjects		37		36			35				100	
between villages		54		51			66				72	

[a]GMR: = geometric mean ratio = GM(estimate)/GM(reference) = exp(Estimate). BaP, benzo[a]pyrene; FLT, fluoranthen; CHR, chrysene; NAP, naphthalene. Example calculation: BaP value for "intercept" home = exp(2.55) = 12.8. BaP value for home using smoky coal from North Xuanwei = exp(2.55 + 1.1) = 38.5, which corresponds to a GMR of 3.01. [b]Reference value represents log transformed PAH value (in ng/m^3) for the reference model entry (smokeless coals from Fuyuan, burnt in a ventilated stove in a "small" room) [c]Refers to smoky coal samples collected from a village in a smokeless coal producing area of Fuyuan.

research on BaP in smoky coal and the classification of BaP as carcinogenic to humans by IARC.[14] The second factor was represented by FLT. This selection was based upon the relative high loading that FLT provides toward the second factor (0.88). CHR was selected to represent the third factor as it was the highest loading variable from that factor (0.58). NAP was selected to represent the gas-phase PAHs as it loaded well toward the single identified factor (0.93) and has been classified as possibly carcinogenic to humans by IARC.[22]

An overview of personal exposure to the selected PAHs is given in Table 2. Particle-bound PAHs (BaP, FLT and CHR) were between 3 and 8 times higher in homes burning smoky coal compared to smokeless coal burning homes ($p < 0.05$). Particle-bound PAH measurements were also observed to be between 4 and 10 times lower among homes burning smoky coal in ventilated stoves when compared to homes burning smoky coal in unvented firepits ($p < 0.05$). Among homes using unventilated stoves, up to a 100 fold difference between smoky and smokeless coal emissions was observed (FLT values of 108.2 and 1.7 ng/m^3 respectively, $p < 0.05$). Gas phase PAHs (NAP) were present in significantly higher concentrations in plant product burning homes (20,000 ng/m^3) than smoky coal burning homes (2,900 ng/m^3, $p < 0.05$). NAP concentrations in smoky coal burning homes using firepits (14 000 ng/m^3) were significantly higher than homes using ventilated stoves (2500 ng/m^3). Descriptive statistics for all PAHs, in addition to calculated BaP equivalent and total PAH values, are available in the SI.

Mixed Effect Modeling. Model construction was aimed at constructing a single, parsimonious model for all four proxy PAHs. Of the approximately 25 variables and combinations of variables considered (SI Table S1), three variables consistently showed a significant effect on all of the measured PAHs with a fourth variable significantly effecting three of the four proxy PAHs. The first variable reflected fuel usage and was entered as a combination of designated fuel types with smoky coal subtypes and their broad geographic source. The second variable reflected stove design, the third variable reflected room volume (divided into quartiles) and the fourth variable represented the season in which measurements were taken (with Spring and Summer merged due to few observations in summer). Descriptive statistics and preliminary analyses indicated that the combustion products of wood and plant products did not significantly vary between different stove designs, therefore, all plant and wood burning homes were considered to be using firepits for the purpose of model construction. An estimate of the strength of effect of each variable (beta (β) effect estimates), 95% confidence intervals and GMR's (GMR = geometric mean ratio = GM(estimate)/GM(reference) = exp(β)) for each selected PAH are available in Table 3.

The model explains 37% of the variance between subjects for BaP, 36% for FLT, 35% for CHR, and 100% for NAP. It also explains 54% of the variance between villages for BaP, 51% for FLT, 66% for CHR, and 72% for NAP.

The use of smoky coal, plant products and wood all result in higher modeled PAH levels than smokeless coal. The use of smoky coal from an unknown source results in the highest GMR for BaP, FLT and CHR (3.42, 5.38, and 6.16 respectively) while the use of meagre lean coal resulted in the highest GMR for NAP (1.93). Among coals with an identified source (i.e., coals which can be directly linked to a producing mine), coking coal from north Xuanwei results in the highest GMR for BaP, FLT, and CHR (3.01, 2.77, and 3.83 respectively). Variation in GMR was observed between the various coal sources. Using BaP as an example of this variation, GMR's ranged between 3.01 (coking coal, north Xuanwei) and 1.26 ($^{1}/_{3}$ coking coal from Fuyuan).

On investigation for the effect of stove design, ventilated stoves resulted in lower PAH exposures than unventilated stoves, portable stoves and firepits. Firepits were associated with the highest GMR's (as compared to ventilated stoves) for BaP, FLT, CHR and NAP (2.94, 5.62, 2.94, and 3.95 respectively).

Room size was found to impact exposure. Larger rooms (those in the third and fourth quartiles) resulted in higher predicted exposure levels for BaP (GMR 1.74 and 1.2 respectively) and FLT (GMR 2.39 and 1.59, respectively) as compared to the first quartile. The second quartile of room size resulted in reduced predicted levels of PAH exposure for BaP, CHR and NAP (GMR 0.83, 0.63, and 0.82 respectively).

Predicted exposure also varied between different seasons. Measurements taken during winter were found to result in the highest predicted levels as compared to measurements taken in autumn (GMR 1.19, 1.86, 1.10, and 1.70 respectively).

■ DISCUSSION

Xuanwei and Fuyuan counties, located in Yunnan province, China have among the highest lung cancer rates in the nation, which is directly associated with the domestic combustion of locally sourced smoky (bituminous) coal.[1−3] Geographic variation in cancer rates[4,9] suggests spatial variation in exposure while a reduction in cancer rates following stove improvement programs[6,7] suggests reduced exposure following improved stove ventilation. Previous research has shown that smoky coal from the region emits high levels of BaP[1,23] when compared to the smokeless (anthracite) variety of coal. Relatively little is known regarding PAH exposure, in particular personal exposure levels in the two counties.

Descriptive statistics show up to an 8 fold difference in particle bound PAH exposure between smoky and smokeless coal. PAH exposure is also up to 10 times lower in smoky coal burning homes using ventilated stoves compared to homes using firepits. These findings parallel previous findings of increased organic material in smoky coal samples,[11] higher levels of overall particulate matter[15] and BaP[23] in smoky coal emissions and reduced BaP exposure with the use of ventilated stoves.[16] No significant difference in gas phase PAHs was observed between smoky coal and smokeless coal. However, up to a 6 fold reduction in gas-phase PAHs was observed among smoky coal burning homes using ventilated stoves compared to firepits.

We note that particle bound PAH measurements among wood and plant burning homes appear to be roughly equivalent to smoky coal burning homes. This finding appears to largely be a feature of stove design. The use of unventilated firepits, is associated with higher particle bound PAH exposure when using smoky coal (GM for BaP, 151.5 ng/m^3) than when using wood (GM, 47.7 ng/m^3), however, in homes using ventilated stoves we observe similar PAH values between wood and smoky coal (GM for BaP 61.2 and 38.1 ng/m^3 respectively). We postulate that this is due to the stoves' designs, which were specifically intended to hold and burn coal. Wood therefore, does not fit properly in these stoves which leads to an underperformance of ventilation and thus little or no reduction in PAH emission. A similar phenomena has also been noted by an IARC working group on HAP.[14] Efficient wood burning stoves are used in some countries. For example, a Tanzanian study observed a reduction of PAH emissions (by approximately 75%) among stoves designed to more efficiently burn wood compared to conventional stoves.[24]

Mixed effect models, created for the purpose of predicting personal PAH exposure levels, provide an expanded view upon the multiple variables which play a role in PAH exposure. The model identifies variations in PAH exposure between differing smoky coal producing areas and differing smoky coal subtypes. Focusing upon smoky coal burning homes which could identify their supplying mine, we observed that coking coal from North Xuanwei (which includes Laibin - the area with the highest lung cancer mortality rate) has the highest GMR (i.e., predicts the highest PAH exposure) when compared to the other smoky coals. The model also predicts reduced PAH exposure of between approximately 3 (BaP, CHR) and 5 (FLT) times between ventilated stoves and firepits. We note that the model indicates that homes with "unknown" stove designs (n = 7) have lower levels of predicted PAH exposure the other stove design categories. These homes represent geographically distinct locations, using a variety of fuel sources from a variety of locations and there is no immediate explanation for this observation. The relationship between relative room size and PAH exposure has not previously been reported. Larger rooms appear to relate to higher PAH exposure, which is unintuitive. A possible explanation, based on field observations, is that smaller rooms indicate financially poorer households, which would have worse construction, allowing for increased general ventilation and reduction in HAP through imperfections in the room's structure. This hypothesis is supported by some of the information collected from the study participants which included a survey of socio-economic indicators. The majority of study participants who reported that they did not regularly have enough to eat (68%) lived in homes in the first and second size quartiles. Conversely, the majority of participants who reported having surplus food (70%) lived in homes in the third and fourth size quartiles. Alternatively, it could be that the larger homes used relatively more fuel. Fuel consumption was accurately recorded during the field survey by weighing the fuel stock pile before and after the measurements. We did not observe a difference in fuel consumption by room size arguing in favor of the fact that in some regards room size reflects social economic status and consequently home design. The role of season in predicted PAH levels, with PAHs being higher during winter, is consistent with expected practices during colder temperatures such as closing windows and spending more time indoors and matches findings of other research which shows increased PAH exposure during colder months.[25] Meteorological indicators (temperature, rainfall, humidity, etc.) were also observed to have a role in predicted levels, however the season category was found to more accurately predict PAH exposure than separate meteorological indicators.

We would not expect tobacco smoking to influence the findings of this study. All of the study enrolees reported that they were not either current or past smokers and over 90% of them reported that at least one of their direct (male) family members regularly smoked. Previous research[26,27] has indicated that HAP attributable to tobacco smoke is less than 1 ng/m^3 thus it is unlikely that the current findings were influenced in any meaningful manner by tobacco smoking.

BaP levels reported here are generally consistent with other research performed in the Xuanwei and Fuyuan counties suggesting that our results are consistent with exposures experienced in the past (these studies typically report the arithmetic mean [AM] of measurements). Lv et.al[28] reported BaP levels of between 7.7 and 380 ng/m^3 (stove design not reported), Tian et.al[16] reported BaP concentrations among unventilated homes burning smoky coal of 901 ng/m^3 and among ventilated homes of 143 ng/m^3, He et.al[2] reported BaP levels of between 286 ng/m^3 and 2485 ng/m^3 for homes burning a variety of solid fuels in unventilated firepits and Lan et al.[7] reported BaP levels of 1660 ng/m^3 among homes burning smoky coal in unvented stoves, compared to 250 ng/m^3 for ventilated stoves. These findings are all substantially lower than those previously reported by Mumford et.al,[23] who reported indoor BaP concentrations of AM 14.7 μg/m^3 (14,700 ng/m^3) in Xuanwei homes burning smoky coal in unvented firepits. The reason for this disparity lays in the fact that Mumford only measured during active cooking periods, which is when exposure is highest. Work by Hosgood et. Al, in the Xuanwei region, has indicated that emission levels can vary by a factor of up to 60 between cooking and noncooking periods.[12] Therefore, measurements taken only during these times are likely to be much higher than those taken over 24 h periods. Of note, if we adjust the AM for BaP resulting from smoky coal being burnt in a fire pit (186.4 ng/m^3) and adjust it for the noted 60-fold difference between cooking and noncooking periods, we would expect exposure levels for BaP of approximately 10 μg/m^3 during peak cooking periods, which is consistent with the reported levels by Mumford et al.

The Chinese national criteria of BaP exposure levels is 1 ng/m^3 for indoor environments,[28] a level which was exceeded by every measurement taken, regardless of fuel type or stove design. The overall AM for BaP concentration among smoky coal homes is 74.4 ng/m^3. In a broader context, this is higher than that observed many other studies throughout China. including coal burning homes from a rural area in northeastern China with a notably high rate of esophageal cancer[29] (39.6 ng/m^3), traffic policemen working in high density Chinese urban environments[30] (26.2 ng/m^3), urban and rural residents of northern China using a variety of fuels[25] (18.9 ng/mg^3) and villagers from northern China using a variety of biomass[31] (24 ng/m^3) . In the international context, we see that BaP levels reported here are relatively high when compared to homes in Poland using coal[32] (28 ng/m^3) in urban India[33] (13.6 ng/m^3) and in coal burning rural Indian homes[34] (56 ng/m^3).

This is the most comprehensive assessment of PAH exposure in Xuanwei and Fuyuan counties to date. These findings parallel observed lung cancer epidemiology in the region. However, risk estimates indicate that the measured PAH concentrations may only account for a 3-fold increase in lung cancer risk,[35−38] which is insufficient to fully explain the lung cancer epidemic in the region. Therefore, it is possible that the risk estimates, which were primarily constructed based on observations of adult men working in coking plants (who would therefore only be exposed during working hours) are inapplicable to women who have been exposed since birth onward. Alternatively, other compounds, either separate to or in combination with PAHs may play an important role in lung cancer etiology.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Supporting Information, containing the variables considered for inclusion in mixed effect model construction; descriptive statistics stratified by fuel type, stove design and fuel source for both indoor and personal measurements; factor analysis of indoor measurements; and mixed effect models for all PAHs, including indoor and personal measurements, is available. 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.

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