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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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 affect 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.4−6 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 rates6,7 (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...
the State Standard of China Coal Classification which recognizes at least four “sub-types” of smoky coal in the region. 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).

Research investigating the properties of uncombusted coal, collected from coal mines and 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. Research investigating the emissions of coal combustion has found that smoky coal (compared to smokeless coal) emits high amounts of nanoparticles, particulate matter, and polycyclic aromatic hydrocarbons (PAHs), specifically benzo[a]pyrene (BaP). BaP is considered a known carcinogen by the International Agency for Research on Cancer (IARC).

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. Additionally, variation in indoor BaP levels has been observed between different geographic locations in Xuanwei, 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, and between 1 and 2 m from the main stove (as allowed by the design of the room).

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 effects. The full details are provided elsewhere 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 enrolles 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 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]fluoranthene (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.
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

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.
content (sum of PAHs) and BaP equivalent (calculated using previously reported toxic equivalency factors for PAHs) 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. PAH limit of detection were imputed from a log-normal probability distribution. Descriptive statistics included on log-transformed PAH values was performed to assess for standard deviations (GSD). ANOVA and Tukey HSD testing on log-transformed values using a log-normal distribution. Factor analysis was performed with varimax rotation.

| Table 1. Exploratory Factor Analysis of Log Transformed Personal PAH Values* |
|-----------------------------------------------------------------------------|
| Factor 1                      | Factor 2                      | Factor 3                      |
| EV(% variance explained)      | EV(% variance explained)      | EV(% variance explained)      |
| benzo[a]pyrene (5)            | 0.81                         | 0.44                         |
| fluorene(4)                   | 0.73                         | 0.60                         |
| pyrene(4)                     | 0.43                         | 0.87                         |
| fluoranthene(4)               | 0.39                         | 0.88                         |
| chrysene(4)                   | 0.56                         | 0.57                         |
| benzo[a]anthracene (4)        | 0.62                         | 0.65                         |
| Factor 1                      | 4.5 (45%)                    | 3.7 (37%)                    |
| Factor 2                      | 3.7 (37%)                    | 1.3 (13%)                    |
| Factor 3                      | 1.3 (13%)                    | 0.29                         |
| gas phase                     |                              |                              |
| naphthalene(2)                | 3.7 (91%)                    |                              |
| phenanthrene (3)              | 0.94                         |                              |
| acenaphthylene (3)            | 0.90                         |                              |
| fluorene (4)                  | 0.99                         |                              |

*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.

Where $y_{ijf}$ represents the natural log transformed value of the PAH exposure levels for village $i$, person $j$ on day $f$. $\mu$ represents the intercept (i.e., the “background” level), $\beta_1$ through $\beta_n$ represent fixed effect variable coefficients for variables $x_1$ through $x_n$, $\gamma_i$ represents the random effect coefficient for village $i$, $b_j$ represents the random effect coefficient for subject $j$, living in village $i$. $\epsilon_{ijf}$ represents the error for village $i$, person $j$ on day $f$.

All statistical testing was carried out in R version 3.03 using the lme 4 package, using $p$ 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.

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
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...
research on BaP in smoky coal and the classification of BaP as carcinogenic to humans by IARC. 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.

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 smoking coal in ventilated stoves when compared to homes burning smoking 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 68.2 and 1.7 ng/m³ respectively, p < 0.05). Gas phase PAHs (NAP) were present in significantly higher concentrations in plant burning homes (20,000 ng/m³) than smokeless coal burning homes (2,900 ng/m³, p < 0.05). NAP concentrations in smoky coal burning homes using firepits (14,000 ng/m³) were significantly higher than homes using ventilated stoves (2500 ng/m³). 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 $\beta$) effect estimates), 95% confidence intervals and GMR’s (GMR = geometric mean ratio = GM(estimate)/GM(reference) = exp($\beta$)) 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 (3.01, 2.77, and 3.83 respectively) while the use of meagre lean coal resulted in lower modeled PAH values for BaP, FLT, and CHR (2.94, 5.62, and 2.94 respectively). The model also predicts reduced PAH exposure when using smoky coal (GM for BaP, 61.2 and 38.1 ng/m3 respectively). The use of通风 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 be a feature of stove design. The use of ventilated firepits, is associated with higher particle bound PAH exposure when using smoky coal (GM for BaP, 151.5 ng/m3) than when using wood (GM, 47.7 ng/m3), 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/m3 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. 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. 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 a higher risk of cancer.

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. Geographic variation in cancer rates suggests spatial variation in exposure while a reduction in cancer rates following stove improvement programs suggests reduced exposure following improved stove ventilation. Previous research has shown that smoky coal from the region emits high levels of BaP 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. This findings parallel previous findings of increased organic material in smoky coal samples, higher levels of overall particulate matter and BaP in smoky coal emissions and reduced BaP exposure with the use of ventilated stoves. 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 ventilated firepits, is associated with higher particle bound PAH exposure when using smoky coal (GM for BaP, 151.5 ng/m3) than when using wood (GM, 47.7 ng/m3), 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/m3 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. 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. 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 a higher risk of cancer.

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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.55 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 enrollees 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 research56,27 has indicated that HAP attributable to tobacco smoke is less than 1 ng/m^3 which is consistent with the reported levels by Mumford et al. This project was supported by the National Institutes of Health (HHSN261201400122P) 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. We acknowledge Johan Beekhuizen and Meng Wang from the Institute for Risk Assessment Sciences, Utrecht University for their support in the production of Figure 1.

The Chinese national criteria for 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 in many other studies throughout China. Including coal burning homes from a rural area in northeastern China with a notably high rate of esophageal cancer29 (39.6 ng/ m^3), traffic policemen working in high density Chinese urban environments30 (26.2 ng/m^3), urban and rural residents of northern China using a variety of fuels35 (18.9 ng/m^3) and villagers from northern China using a variety of biomass31 (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 coal32 (28 ng/m^3) in urban India33 (13.6 ng/m^3) and in coal burning rural Indian homes34 (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

5 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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The authors declare no competing financial interest.

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