Published in final edited form as:

Indoor Air. 2016 October ; 26(5): 784–795. doi:10.1111/ina.12255.

Outdoor, indoor, and personal black carbon exposure from cookstoves burning solid fuels

George S. Downward^{1,†}, Wei Hu², Nat Rothman², Boris Reiss¹, Guoping Wu³, Fusheng Wei³, Jun Xu⁴, Wei Jie Seow², Bert Brunekreef¹, Robert S. Chapman⁵, Lan Qing^{2,*}, and Roel Vermeulen^{1,*}

¹Institute for Risk Assessment Sciences, Division of Environmental Epidemiology, Utrecht University, Utrecht, The Netherlands

²Division of Cancer Epidemiology and Genetics, National Cancer Institute, NIH, DHHS, Bethesda, MD, USA

³China National Environmental Monitoring Centre, Beijing, China

⁴Hong Kong University, Hong Kong, China

⁵College of Public Health Sciences, Chulalongkorn University, Bangkok, Thailand

Abstract

Background—Black carbon (BC) emissions from solid fuel combustion are associated with increased morbidity and mortality and are important drivers of climate change. We studied BC measurements, approximated by particulate matter (PM_{2.5}) absorbance, in rural Yunnan province, China whose residents use a variety of solid fuels for cooking and heating including: bituminous and anthracite coal, and wood.

Methods—Measurements were taken over 2 consecutive 24 h periods from 163 households in 30 villages. PM_{2.5} absorbance (PM_{abs}) was measured using an EEL 043 Smoke Stain Reflectometer.

Results—PM_{abs} measurements were higher in wood burning households ($16.3 \times 10^{-5} \text{ m}^{-1}$) than bituminous and anthracite coal households (12 and $5.1 \times 10^{-5} \text{ m}^{-1}$ respectively). Among bituminous coal users, measurements varied by a factor of two depending on the coal source. Portable stoves (which are lit outdoors and brought indoors for use) were associated with reduced PM_{abs} levels, but no other impact of stove design was observed. Outdoor measurements were positively correlated with and approximately half the level of indoor measurements ($r = 0.49$, $p < 0.01$).

Conclusion—Measurements of BC (as approximated by PM_{abs}) in this population are modulated by fuel type and source. This provides valuable insight into potential morbidity, mortality and climate change contributions of domestic usage of solid fuels.

[†]Corresponding author, Correspondence: George Downward, Institute for Risk Assessment Sciences, Utrecht University, Utrecht, Yalelaan 2, 3584CM, The Netherlands, Phone: +31 30 253 2578, g.s.downward@uu.nl.

*These authors co-supervised this work.

Competing financial interests declaration

The authors declare that there is no competing financial interest in the planning, analysis or publication of this research.

Keywords

Black carbon; Coal; China; Solid fuels; Household air pollution; Climate change; PM_{2.5} absorbance

1. Introduction

Xuanwei, and its neighbouring county of Fuyuan, located in Yunnan province, China have among the highest lung cancer rates in the nation, regardless of gender or smoking status (Barone-Adesi et al., 2012; Mumford et al., 1987). The residents of Xuanwei and Fuyuan, like approximately 3 billion people world-wide, are largely dependent upon the use of solid fuels (coal, wood etc.) for their daily heating and cooking needs (World Health Organization, 2006), a practise which annually accounts for up to 4 million deaths world-wide (Gordon et al., 2014). Locally sourced “smoky” coal (a bituminous coal) is the primary fuel source mined and used throughout the Xuanwei and Fuyuan counties (Figure 1) and has, through multiple lines of research, been identified as the cause of the excess lung cancer mortality in the region (Barone-Adesi et al., 2012; Chapman et al., 1988; He et al., 1991; Mumford et al., 1987). Approximately 25% of residents use alternative fuels, including “smokeless” coal (an anthracite coal available in a few locations), wood, corn cobs, and tobacco stems.

Previous research has identified that household air pollution (HAP) related to smoky coal use (when compared to smokeless coal) contains relatively high amounts of particulate matter (PM_{2.5}) and polycyclic aromatic hydrocarbons (PAH) (Downward et al., 2014b; Hu et al., 2014). However, exposure to these compounds alone appears to be insufficient to explain the lung cancer risk in the area, requiring the consideration of alternative pollutants. Thus far, nothing is known regarding the role that black carbon (BC) plays in the area. Black carbon is released via the incomplete combustion of solid and fossil fuels and is an important predictor of mortality and other adverse health effects in studies of ambient air pollution (Bond et al., 2004; Grahame et al., 2014). There is also a suggestion that in some settings it provides a superior estimate of morbidity and mortality than PM_{2.5}. For example, in a meta-analysis of time series studies, Janssen et. al. found that exposure to BC was associated with an increase in all-cause mortality and that the estimated health effect for increased BC exposure was greater than that for particulate mass. (Janssen et al., 2011). Black carbon exposure has also been associated with increased blood pressure, type 2 diabetes, and respiratory infections among children under 36 months of age (Baumgartner et al., 2014; Krämer et al., 2010; Macintyre et al., 2013).

An additional feature of BC is that by absorbing heat and reducing albedo (reflected sunlight) it is an important climate change driver (Anenberg, 2012; Martin et al., 2014; Schmidt, 2011). Unlike carbon dioxide, BC only remains in the atmosphere for a short time (days to several weeks) (United Nations Environment Programme and World Meteorological Organization, 2011), meaning that reductions in BC emissions will have an immediate impact upon climate forcing. Up to 80% of BC emissions in Africa and Asia have been attributed to residential solid fuel use (Bond et al., 2013) and there is evidence to suggest

that improved stove designs can significantly reduce BC emissions. A study in Indian households found up to a 90% reduction in indoor BC emissions if stoves with forced ventilation (i.e. fan driven chimney) were utilized (Kar et al., 2012). In Xuanwei and Fuyuan, solid fuels were historically burnt in unvented firepits while recent decades have seen the implementation of stove improvements, particularly the installation of fixed stoves attached to chimneys, aimed at increasing burning efficiency and passively venting fuel emissions outdoors. These improved stoves have been associated with reduced PM_{2.5} and PAH air concentrations (Downward et al., 2014b; Hu et al., 2014; Tian et al., 2009) in addition to reductions in both malignant and non-malignant lung disease (Chapman et al., 2005; Hosgood et al., 2008; Lan et al., 2002; Lee et al., 2010; Shen et al., 2009). However, as the stove improvements carried out in Xuanwei and Fuyuan were focussed upon the installation of fixed cook-stoves with attached chimneys and did not include fan-forced ventilation, it is unclear whether a reduction in indoor BC levels would be achieved. Furthermore, the consequences for the general outdoor air quality are unknown.

Given the importance of BC, in terms of both a health assessment and climate change perspective, there is value in understanding the relationship between solid fuel use and BC measurements in Xuanwei and Fuyuan. This paper will present personal, indoor, and outdoor BC measurements and explore the various roles that fuel type, fuel source and stove design play on measured concentrations.

2. Methods

Data for this paper was collected as part of a large cross-sectional molecular epidemiology study aimed at cataloguing the constituents of solid fuels and their associated air pollution from throughout Xuanwei and Fuyuan and associating those constituents with biological effect markers and lung cancer risk in a case control study of lung cancer among never-smoking females. The full details are provided elsewhere (Downward et al., 2014a) but briefly, 30 villages were selected from throughout Xuanwei and Fuyuan (15 villages from each county). Approximately 5 households with a healthy non-smoking adult female resident were selected from each village. Household selection was directed towards reflecting the population in the case control study, which represents fuel and stove usage for the previous several decades. Therefore houses which were at least 10 years old and had not had any stove alterations undertaken in the past 5 years were preferentially selected. One non-smoking female, between the ages of 20 and 80 who resided in the household and was primarily responsible for cooking, was enrolled for personal monitoring of pollutants. Written informed consent was gained prior to enrolment.

Homes were measured and sketched. Pertinent features (e.g. stoves, doors, windows and stairways) were recorded. Data was collected during 2 collection periods. The first was between August 2008 and February 2009. During this period 148 participants were recruited from all 30 villages. During the second collection period, between March and June 2009, 16 of the original 30 villages were re-visited (villages were selected based upon how well the stove and fuel use recorded in phase 1 represented the overall study population), 53 of the original households and enrolees were re-visited and 15 new subjects were enrolled. During each collection period, measurements were taken during 2 consecutive 24 hour periods.

Following a review of reported fuel types, the following categories for fuel type were identified: smoky coal, smokeless coal, “other” coal (combinations of smoky coal, smokeless coal and processed coal products such as briquettes), wood, plant products (combinations of corn cobs, tobacco stems and bamboo shoots, sometimes in combination with wood) and “other” fuels (combinations of coal and plants). When a subject reported using coal, geochemical analysis was used to confirm coal type (Downward et al., 2014a). Subsequently, smoky coal sub-types were classified according to the Chinese State Standard for coal classification (the subtypes are: coking coal, 1/3 coking coal, gas fat coal and meagre lean coal [Figure 1]) (Chen, 2000).

Stoves were categorized based upon their ventilation design with the following categories used for analysis: ventilated stoves, unventilated stoves, firepit, portable stove (which is designed to be lit outside and carried indoors for use), “mixed” (which refers to the use of multiple stoves some with, and some without chimneys) and unknown (where stove type was not recorded).

2.1 Sample collection

Black carbon was measured through the commonly used proxy measurement of light absorbance of the fine particulate matter (PM_{abs}) (Cyrys et al., 2003). Personal and indoor $PM_{2.5}$ measurements were collected on 37 mm Teflon filters attached to a cyclone with an aerodynamic cut-off of $2.5 \mu m$ (model BGI, GK 2.05SH) at a flow rate of 3.5 L/min ($\pm 20\%$). Pumps underwent calibration prior to all measurements and flow rates were recorded pre and post sampling. If the post-sampling flow deviated by more than 10%, data was not accepted for further analysis (approximately 2% of filters were considered not acceptable for use). Filters were changed after approximately 24 hours of operation (median period: 23.0 hours, IQR: 22.0 – 23.6 hours). For personal measurements, the pump was packed in a hip bag and the cyclone was attached near the breathing zone in order to approximate personal exposure. Overnight, the equipment was put next to the subject’s bed. Indoor measurements were collected using the same methods as described above with samplers being placed in the main living area. Samplers were placed at least 0.25 m from each wall and 1–2 m from stoves (as allowed by indoor space). If there was a separate room which contained a stove for cooking or heating, then an additional indoor measurement was taken in that room (this represents 6% of indoor measurements).

Outdoor PM_{abs} measurements were taken at a central location in each village away from any direct local pollution sources (such as chimneys). External sources of pollution (factories, power plants etc.) within 5 km of each village were documented. The main fuels and stoves in use by inhabitants of each village was established by accessing local records, collected by local health professionals which contained overall fuel and stove usage for each village.

After sample collection, all exposed filters were individually packed into petri slides and stored in zipped amber plastic bags while in transit for analysis.

2.2 Sample analysis

Light reflectance was measured across 5 different spots on the filter (to compensate for variation across the filter) using an EEL 043 Smoke Stain Reflectometer. The average

reflectance across the 5 points was transformed into an absorption value using the following formula (ISO, 1993).

$$a = \frac{A}{2V} \ln \left(\frac{R_0}{R_f} \right) \times 100,000. \quad (\text{Equation 1})$$

Where a represents the absorption coefficient (in 10^{-5}m^{-1}), A the area of the loaded filters (0.00078 m^2), V the volume of sampled air (in m^3), R_0 the average reflectance of field blanks (102%) and R_f the reflectance of the sampled filter (%).

2.3 Quality Control

As stated above, all samples were blank corrected. Reflectance of filters was completed over several measurement “batches”. During each “batch”, approximately 10% of filters in that batch had their measurements repeated. If the average reflectance of the repeat measurement deviated by more than 3%, all filters being measured were re-measured. Additionally, field duplicates were taken for approximately 25% of all samples ($n=211$ [203 indoor and 8 personal measurements]). The median relative percentage difference of these repeat measurements was 7% (Interquartile range [IQR] 3.4 to 13.5%).

2.4 Comparison to other components of air pollution

Measurements of $\text{PM}_{2.5}$ and PAH have been described previously (Downward et al., 2014b; Hu et al., 2014). By matching $\text{PM}_{2.5}$ and PAH measurements to absorbance measurements collected on the same day, we assessed the interrelationship between PM_{abs} and other air pollution components.

2.5 Statistical Analysis

Normal probability plots indicated that measured absorbance values tended towards log-normality. Descriptive statistics include arithmetic means (AM), geometric means (GM) and geometric standard deviations (GSD). Analysis of variance (ANOVA) and Tukey honestly significant difference (HSD) testing, carried out on log-transformed values, was used to assess for differences between fuel types, stove designs and smoky coal sources. When assessing between smoky coal sources, only smoky coal which could be linked to a specific coal mine was included for analysis.

In order to further explore the factors influencing absorbance measurements, linear mixed effect models (on the log transformed values) were constructed. In the construction of models, individual subjects, and the villages that they resided in were assigned as random effects. Approximately 25 variables (see Table S1 for a full list of considered variables), including fuel source, stove design, season, house characteristics (e.g. size, number of windows, and presence or absence of a stairway) and meteorological factors were considered for inclusion as fixed effects. Inclusion of variables in the final model was based on the combination of how they influenced the Akaike information criterion (AIC) score and how they best described the observed data. The model can be represented with the following formula:

$$y_{ijf} = \mu + \beta_1 x_1 + \beta_2 x_2 \dots \beta_n x_n + bI_i + bJ_{ij} + \varepsilon_{ijf} \quad (\text{Equation 2})$$

Where y_{ijf} represents the natural log transformed value of absorbance measurements 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 ; bI_i represents the random effect coefficient for village i ; bJ_{ij} represents the random effect coefficient for subject j , living in village i ; and ε_{ijf} represents the error for village i , person j on day f .

All statistical testing was carried out using R version 3.0.2 (R Development Core Team, 2014) using the lme4 package (Bates et al., 2014). A p value of less than 0.05 was considered to indicate statistical significance.

3. Results

In total, 923 absorbance measurements were collected (414 personal, 443 indoor, and 66 village based measurements). Of these measurements, 411 (202 personal and 209 indoor) represented the exclusive use of smoky coal, 398 of which (195 personal and 203 indoor) could be assigned to individual mines. In Xuanwei, all smoky coal producing mines reported were of the coking coal subtype while in Fuyuan smoky coal mines spanned a variety of subtypes: coking coal, 1/3 coking coal, gas fat coal and meagre lean coal. Among village-based samples, 38 samples were collected from villages where smoky coal was the dominant fuel used, 10 smokeless, and 18 samples were from villages where there was no clear dominant fuel type used.

3.1 Personal and Indoor measurements

Indoor and personal measurements correlated well with each other, with a Spearman correlation coefficient of 0.70 ($p < 0.01$). In general, personal measurements were similar to indoor (median percentage difference: +6.6% IQR: -21.7% to +26.9%). A mixed effect model, using the approach described in Equation 2, was constructed to explore variables which may contribute to the observed difference between indoor and personal measurements (Table S2 in the supplement) which indicated that personal measurements would be higher (relative to indoor measurements) during colder temperatures, during periods when more fuel (in kg) was used and during periods of extended stove usage.

An overview of $PM_{2.5}$ absorbance is shown in Table 1. Households burning wood had the highest PM_{abs} for both personal ($16.3 \times 10^{-5} m^{-1}$) and indoor ($15.0 \times 10^{-5} m^{-1}$) measurements and for personal measurements they were significantly higher than households burning smoky coal ($12 \times 10^{-5} m^{-1}$, $p < 0.05$). Households exclusively burning smoky coal had significantly higher PM_{abs} than households burning smokeless coal for both personal ($12 \times 10^{-5} m^{-1}$ vs $5.1 \times 10^{-5} m^{-1}$, $p < 0.05$) and indoor ($11.8 \times 10^{-5} m^{-1}$ vs $4.7 \times 10^{-5} m^{-1}$, $p < 0.05$) measurements. On investigation for the role of stove design, households burning smoky coal in portable stoves had significantly lower personal and indoor measurements than those using unventilated stoves (8.9 and $7.7 \times 10^{-5} m^{-1}$ vs 12.0 and 12.2

$\times 10^{-5} \text{ m}^{-1}$ respectively, $p < 0.05$). No other significant difference between stove designs (e.g. presence or absence of a chimney) was observed.

On assessing for variation between different smoky coal sources (Table 2), gas fat coal from the Fuyuan mine of “Qingyun” had the highest personal and indoor PM_{abs} values ($23.7 \times 10^{-5} \text{ m}^{-1}$ and $21.3 \times 10^{-5} \text{ m}^{-1}$ respectively) while the coking coal mine “Daping” also located in Fuyuan had the lowest ($6.8 \times 10^{-5} \text{ m}^{-1}$ and $6.0 \times 10^{-5} \text{ m}^{-1}$). ANOVA testing revealed significant variation within coking coals sourced from within Xuanwei (range for personal measurements $10.7 \times 10^{-5} \text{ m}^{-1}$ to $16.4 \times 10^{-5} \text{ m}^{-1}$, $p < 0.05$), within the smoky coal subtypes produced in Fuyuan (range $5.2 \times 10^{-5} \text{ m}^{-1}$ for meagre lean coal to $13.2 \times 10^{-5} \text{ m}^{-1}$ for gas fat coal) and within the Fuyuan coking coals ($6.8 \times 10^{-5} \text{ m}^{-1}$ to $14.7 \times 10^{-5} \text{ m}^{-1}$), 1/3 coking coals ($7.1 \times 10^{-5} \text{ m}^{-1}$ to $11.6 \times 10^{-5} \text{ m}^{-1}$, personal measurements only) and the gas fat coal subtypes ($11.7 \times 10^{-5} \text{ m}^{-1}$ to $21.3 \times 10^{-5} \text{ m}^{-1}$, indoor measurements only).

3.1.1 Mixed effect modelling—Construction of a linear mixed effect model indicated that fuel type and source, stove design, the season of measurements and the presence or absence of a stairway all contributed to PM_{abs} values. An estimate of the strength of effect of each variable (beta (β) effect estimates), 95% confidence intervals and GMR's (GMR = geometric mean ratio = $\text{GM}(\text{estimate})/\text{GM}(\text{reference}) = \exp(\beta)$) are available in Table 3. The model explains 17% of the variance between subjects and 66% of the variance between villages for personal measurements (28% and 47% respectively for indoor measurements). It indicates that the use of wood as a fuel source results (when compared to smokeless coal) in the highest PM_{abs} values (GMR 2.22 for personal and 1.67 for indoor measurements). Variation in PM_{abs} values was observed between the differing smoky coal types and sources (e.g. among personal measurements the GMR ranges from 0.92 for meagre lean coal to 1.79 for gas fat coal). The exclusive use of a portable stove (compared to a ventilated stove) resulted in the lowest predicted PM_{abs} values (GMR 0.84 for both indoor and personal measurements). The presence or absence of a stairway in the main cooking room also impacted PM_{abs} values. Homes without stairways had lower PM_{abs} than those with them (GMR 0.88 for both indoor and personal measurements). Measurements taken during winter (when compared to autumn) resulted in the highest PM_{abs} (GMR 1.32 for personal measurements and 1.19 for indoor measurements) while those taken in summer resulted in the lowest (GMR 0.85 for personal and 0.77 for indoor measurements).

3.1.2 Comparisons to other air measurements—Previous research on this population has presented personal and indoor particulate mass ($\text{PM}_{2.5}$) measurements (Hu et al., 2014). In brief, this study found that, similarly to the current study, $\text{PM}_{2.5}$ measurements were higher among smoky coal users ($148 \mu\text{g}/\text{m}^3$) than smokeless ($115 \mu\text{g}/\text{m}^3$) but lower than wood users ($289 \mu\text{g}/\text{m}^3$). Unlike the current study, $\text{PM}_{2.5}$ measurements were lower among users of ventilated stoves than users of firepits (values for smoky coal users: $150 \mu\text{g}/\text{m}^3$ for vented stoves and $307 \mu\text{g}/\text{m}^3$ for firepits, $p < 0.05$). The ratio of PM_{abs} to $\text{PM}_{2.5}$ for smoky coal burnt in a firepit was 1:25 while for smoky coal in a vented stove it was 1:10, which is likely illustrative of the differing roles that stove design play in these modalities. Despite the differing exposure patterns between stove types, PM_{abs} and $\text{PM}_{2.5}$ measurements showed a strong degree of correlation with each other (Spearman r : 0.68, $p < 0.05$).

Measurements of polycyclic aromatic hydrocarbons (PAH) have also been presented for this population (Downward et al., 2014b). Using Benzo[a]Pyrene (BaP) as an example of the measured PAHs, measurements were higher among smoky coal users than smokeless (44.7 ng/m^3 vs 10.6 ng/m^3 , $p < 0.05$) and smoky coal burnt in a ventilated stove was associated with lower BaP measurements than if it was burnt in a firepit (38.1 ng/m^3 vs 151.5 ng/m^3 , $p < 0.05$). There was also a strong correlation between BaP and PM_{abs} measurements ($r: 0.73$, $p < 0.05$). Similarly to the $\text{PM}_{2.5}$ measurements which in itself correlates with BaP ($r: 0.73$, $p < 0.05$), a major source of the relative difference between PAH and PM_{abs} measurements was stove design. The $\text{PM}_{\text{abs}}:\text{BaP}$ ratio for smoky coal burnt in a firepit was 1:11 while for smoky coal in a vented stove it was 1:3.

3.2 Village based outdoor measurements

Village based outdoor measurements correlated moderately well with both the average indoor and personal measurements of study enrolees per village (Spearman correlation coefficients 0.43 and 0.49 respectively, $p < 0.05$ for both measurement types [Figure S1 in supplemental material]). In general, village based measurements were lower than both indoor and personal measurements (median percent difference -54% [IQR: -66% to -45%] and -57% [IQR: -67% to -49%] respectively).

Village based PM_{abs} measurements were highest among villages where smoky coal was the dominant fuel used (GM: $5.7 \times 10^{-5} \text{ m}^{-1}$) and lowest among smokeless coal using villages (GM: $3.1 \times 10^{-5} \text{ m}^{-1}$, p value for difference < 0.05 , Table 4). There was no significant variation observed in outdoor PM_{abs} values between the stove types used in the villages. Investigation for variation among smoky coal sources showed no significant variation in PM_{abs} values. On assessment for the role of external pollutant sources, there was a weak but statistically significant correlation between the total number of pollution sources (e.g. factories, power plants) and PM_{abs} (Spearman correlation coefficient: 0.28, $p < 0.05$).

Construction of a linear mixed effect model indicated that log-transformed indoor absorbance measurements, the average outdoor relative humidity and the season of measurement contributed to outdoor absorbance levels. Effect estimates (β), 95% confidence intervals and GMR's are available in Table 5.

The model explains 52% of the variance between villages and indicates that outdoor absorbance measurements are highest during winter (GMR 1.41). Increasing indoor PM_{abs} values result in an increase in outdoor measurements and increasing outdoor humidity results in a decrease in outdoor absorbance measurements. The number of external pollution sources did not contribute to the final model.

4. Discussion

Black carbon (BC) emissions from the domestic combustion of solid fuels has important ramifications for both human health and global climate change (Anenberg, 2012; Martin et al., 2014; Schmidt, 2011). This is the first study to investigate BC, as approximated by $\text{PM}_{2.5}$ absorbance (PM_{abs}), in relation to the combustion of solid fuels in the Chinese counties of Xuanwei and Fuyuan, where the combustion of coal is associated with multiple

health complications, including the highest lung cancer rates in the nation (Barone-Adesi et al., 2012; Mumford et al., 1987).

Enrolees burning wood were found to have the highest personal PM_{abs} levels ($16.3 \times 10^{-5} m^{-1}$) followed by plant and smoky coal (12.5 and $12.0 \times 10^{-5} m^{-1}$ respectively). This indicates that BC exposure alone is likely insufficient to explain the lung cancer epidemic in the area (as smoky coal which carries the highest lung cancer risk, would therefore be expected to be associated with the highest PM_{abs} measurements). This is supported further by the observation that PM_{abs} values are not affected by the type of stove used (with the exception of portable stoves) which is contrary to previous findings of reduced lung cancer rates following the installation of stoves with chimneys (Lan et al., 2002). Despite these findings, BC exposure in the area is still of significant public health concern as the PM_{abs} measurements reported in the current study are high when compared to other studies. Our personal/indoor PM_{abs} levels are up to 15 times higher than those measured in western settings. For example, in a study investigating personal exposure to air pollution among coronary patients in Amsterdam and Helsinki, geometric means of 1.33 and $1.26 \times 10^{-5} m^{-1}$ are reported (Lanki et al., 2007). Another study, investigating indoor measurements of PM_{abs} in homes in the Bronx and northern Manhattan, New York during cold months reported indoor PM_{abs} measurements of (arithmetic mean) $0.97 \times 10^{-5} m^{-1}$ (Jung et al., 2010).

High PM_{abs} measurements are not only observed indoors. Xuanwei and Fuyuan are primarily rural areas, with less than half of enrolled villages having an identified external pollution source (e.g. a factory), but the average outdoor absorbance measurements reported in the current study ($6 \times 10^{-5} m^{-1}$) are frequently in excess of those by a European urban roadside. For example, several large multi-centre studies exploring air pollution throughout Europe, have reported average absorbance concentrations of less than $2 \times 10^{-5} m^{-1}$ (Durant et al., 2014; Eeftens et al., 2012).

By converting measured absorbance values to a mass concentration ($\mu g/m^3$) it is possible to compare measurements reported in the current study to a wider variety of literature. This conversion is based upon calculations performed by Quincy and converts absorbance to a concentration of black carbon, using a conversion to black smoke as an intermediate step (Quincey, 2007). This conversion results in an average personal black carbon measurement of (GM) $18 \mu g/m^3$ (AM $18.6 \mu g/m^3$) among users of wood, $14 \mu g/m^3$ (AM $16.6 \mu g/m^3$) among smoky coal users and $6.2 \mu g/m^3$ (AM: $7.2 \mu g/m^3$) among smokeless coal users. Compared to other studies, these values are relatively high. For example, these values are higher than personal measurements reported in western Yunnan ($5.2 \mu g/m^3$) and in rural Sichuan ($3.2 \mu g/m^3$). Both of these communities also rely on solid fuels for their domestic tasks (Baumgartner et al., 2014; Shan et al., 2014). Furthermore, wood and smoky coal measurements are also higher than those reported for personal exposure to black carbon among users of biomass in Ghana (AM: $8.8 \mu g/m^3$) (Van Vliet et al., 2013).

A global strategy to mitigate the health and climate change impacts of solid fuel use has been cook-stove improvement, allowing for a more efficient use of fuel, resulting in a reduction in BC emissions (United Nations Environment Programme and World Meteorological Organization, 2011). In Xuanwei and Fuyuan, the installation of ventilated

stoves has resulted in reductions in both malignant and non-malignant lung disease, which has been reflected by reductions in personal and indoor measurement of pollutants (Chapman et al., 2005; Downward et al., 2014b; Hosgood et al., 2008; Hu et al., 2014; Lan et al., 2002; Shen et al., 2009). The finding that ventilated stoves (i.e. stoves with chimneys) in Xuanwei and Fuyuan do not result in significantly reduced PM_{abs} when compared to unventilated stoves and firepits is contrary to these previous findings. However, they mirror those reported in an Indian study (Kar et al., 2012) which reported that stoves operating without some form of forced ventilation (e.g. a fan driven chimney) showed no difference in indoor BC emissions between unventilated cookstoves and those with un-forced ventilation. This indicates that, in order to properly mitigate contributions to climate change and minimize local morbidity and mortality, further stove improvements, or preferably, replacing solid fuel stoves with alternative cooking and heating sources (e.g. biogas or electric stoves) are warranted.

The reduction in PM_{abs} measurements among households using portable stoves is likely a reflection of how these stoves are operated. Portable stoves are designed to be lit outdoors and only brought indoors after the initial ignition period, which is the period during which particulate emissions are maximal (Hosgood et al., 2012). As the stove is moved away from the indoor measurement devices, and residents leave the stoves unattended while outdoors, it therefore follows, that indoor and personal measurements will be lower.

We note also that one of the outputs of the mixed effect model for personal and indoor PM_{abs} values exposure was that homes without a stairway had lower predicted values than homes with one. This appears counter-intuitive as it would otherwise be expected that the presence of a stairway could reduce measurements as a result of increasing the overall circulating volume. A possible explanation is that having a stairway is an indicator of economic status and home construction. Based upon field observations, poorer households are likely to be smaller and less likely to possess stairways and are less structurally sound, resulting in increased air flow of the rooms through structural imperfections. Socio-economic information gathered from study participants appears to support this hypothesis. Approximately 86% of people living in homes without stairways reported either having “not enough” or “just enough” food to eat, compared to 65% of people living in homes with stairways.

It is important to note that many of the health impacts associated with BC exposure have been derived from studies in which the main source of BC is combustion engines (i.e. diesel) and not solid fuel cookstoves. Therefore, due to the heterogeneous nature of BC constituents, it is possible that the health impacts of BC derived from diesel combustion may not directly translate to equivalent health risks for BC derived from solid fuel combustion. We also note that the situation in Xuanwei and Fuyuan is unique and that the coals used in the region may not be constitutionally similar to other coals of the same rank (i.e. bituminous, anthracite) sourced in other areas. Therefore, the results of the current study may be of limited generalizability to other coal-using regions. Additionally, there is a danger that at high concentrations of particulate matter, the degree of filter loading is too high to reliably identify higher absorbance values. An assessment of the relationship between $PM_{2.5}$ and PM_{abs} revealed that at high $PM_{2.5}$ concentrations (over approximately $150 \mu g/m^3$) there

may be some attenuation in PM_{abs} measurements. The consequence of this is that higher absorbance measurements may be under-estimated. To assess any potential impact of this under-estimation on the main findings of the current paper, two separate sensitivity analyses were conducted. First, we analysed absorbance measurements at $PM_{2.5}$ values below $150 \mu g/m^3$. This analysis found similar relative differences between fuel and stove types as what has been presented in the current paper. The second sensitivity analysis involved the creation of a mixed effect model, based upon the relationship between PM_{abs} and $PM_{2.5}$ at $PM_{2.5}$ concentrations below $150 \mu g/m^3$ and using that relationship to model absorbance measurements at $PM_{2.5}$ values above $150 \mu g/m^3$. The corrected PM_{abs} were slightly higher than those recorded (median percentage difference: 5%) however the relationship between fuel and stove types remained unchanged. On the strength of these sensitivity analyses we are confident that even if an underestimation of PM_{abs} has occurred at higher $PM_{2.5}$ levels that this underestimation appears to be relatively small and the main findings and conclusions of this paper (specifically the roles that fuel and stove types play in measurements) are robust.

4.1 Summary and Conclusion

The use of solid fuels in Xuanwei and Fuyuan (especially smoky coal) is related to high amounts of both malignant and non-malignant lung disease. Indoor and personal measurements of black carbon, as measured by the proxy of $PM_{2.5}$ absorbance, are in excess of both urban roadsides and other communities burning solid fuels. This is of significant concern from both a climate change and public health perspective. The ventilated stove designs currently used in Xuanwei and Fuyuan, which have previously been associated with reduced malignant and non-malignant respiratory disease in addition to reductions in $PM_{2.5}$ and PAH emissions do not appear to be associated with reduced PM_{abs} , indicating that further stove improvements would be required before health and climate effects of BC production would be mitigated. As PM_{abs} values from wood burning is greater than that of smoky coal, it is likely that even though exposure to BC is likely to be of significant public health concern, it alone is not sufficient to explain the lung cancer epidemic in the region and that investigation of other constituents of household air pollution is warranted.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements

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. The authors would like to acknowledge Johan Beekhuizen and Meng Wang from the Institute for Risk Assessment Sciences, Utrecht University for their support in the production of figure 1.

References

Anenberg S. Technology: clean stoves benefit climate and health. *Nature*. 2012; 490:343. [PubMed: 23075974]

- Barone-Adesi F, Chapman RS, Silverman DT, He X, Hu W, Vermeulen R, Ning B, Fraumeni JF Jr, Rothman N, Lan Q. Risk of lung cancer associated with domestic use of coal in Xuanwei, China: retrospective cohort study. *BMJ*. 2012; 345:e5414. [PubMed: 22936785]
- Bates D, Maechler M, Bolker B, Walker S. lme4: linear mixed-effects models using Eigen and S4. 2014
- Baumgartner J, Zhang Y, Schauer JJ, Huang W, Wang Y, Ezzati M. Highway proximity and black carbon from cookstoves as a risk factor for higher blood pressure in rural China. *Proceedings of the National Academy of Sciences*. 2014; 111:13229–13234.
- Bond TC, Doherty SJ, Fahey DW, Forster PM, Bernsten T, DeAngelo BJ, Flanner MG, Ghan S, Kärcher B, Koch D, Kinne S, Kondo Y, Quinn PK, Sarofim MC, Schultz MG, Schulz M, Venkataraman C, Zhang H, Zhang S, Bellouin N, Guttikunda SK, Hopke PK, Jacobson MZ, Kaiser JW, Klimont Z, Lohmann U, Schwarz JP, Shindell D, Storelvmo T, Warren SG, Zender CS. Bounding the role of black carbon in the climate system: a scientific assessment. *Journal of Geophysical Research: Atmospheres*. 2013; 118:5380–5552.
- Bond TC, Streets DG, Yarber KF, Nelson SM, Woo J, Klimont Z. A technology-based global inventory of black and organic carbon emissions from combustion. *Journal of Geophysical Research: Atmospheres*. 2004; 109:D14203.
- Chapman RS, He X, Blair AE, Lan Q. Improvement in household stoves and risk of chronic obstructive pulmonary disease in Xuanwei, China: retrospective cohort study. *BMJ*. 2005; 331:1050. [PubMed: 16234255]
- Chapman RS, Mumford JL, Harris DB, He ZZ, Jiang WZ, Yang RD. The epidemiology of lung cancer in Xuan Wei, China: current progress, issues, and research strategies. *Arch Environ Health*. 1988; 43:180–185. [PubMed: 3377554]
- Chen P. Study on integrated classification system for Chinese coal. *Fuel Process Technol*. 2000; 62:77–87.
- Cyrys J, Heinrich J, Hoek G, Meliefste K, Lewné M, Gehring U, Bellander T, Fischer P, Vliet Pv, Brauer M, Wichmann H, Brunekreef B. Comparison between different traffic-related particle indicators: elemental carbon (EC), PM_{2.5} mass, and absorbance. *J Expo Anal Environ Epidemiol*. 2003; 13:134–143. [PubMed: 12679793]
- Downward GS, Hu W, Large D, Veld H, Xu J, Reiss B, Wu G, Wei F, Chapman RS, Rothman N, Qing L, Vermeulen R. Heterogeneity in coal composition and implications for lung cancer risk in Xuanwei and Fuyuan counties, China. *Environ Int*. 2014a; 68:94–104. [PubMed: 24721117]
- Downward GS, Hu W, Rothman N, Reiss B, Wu G, Wei F, Chapman RS, Portengen L, Qing L, Vermeulen R. Polycyclic aromatic hydrocarbon exposure in household air pollution from solid fuel combustion among the female population of Xuanwei and Fuyuan counties, China. *Environ Sci Technol*. 2014b; 48:14632–14641. [PubMed: 25393345]
- Durant JL, Beelen R, Eeftens M, Meliefste K, Cyrys J, Heinrich J, Bellander T, Lewné M, Brunekreef B, Hoek G. Comparison of ambient airborne PM_{2.5}, PM_{2.5} absorbance and nitrogen dioxide ratios measured in 1999 and 2009 in three areas in Europe. *Sci Total Environ*. 2014; 487:290–298. [PubMed: 24793326]
- Eeftens M, Tsai M, Ampe C, Anwander B, Beelen R, Bellander T, Cesaroni G, Cirach M, Cyrys J, de Hoogh K, De Nazelle A, de Vocht F, Declercq C, Dedele A, Eriksen K, Galassi C, Gražulevičienė R, Grivas G, Heinrich J, Hoffmann B, Iakovides M, Ineichen A, Katsouyanni K, Korek M, Krämer U, Kuhlbusch T, Lanki T, Madsen C, Meliefste K, Mölter A, Mosler G, Nieuwenhuijsen M, Oldenwening M, Pennanen A, Probst-Hensch N, Quass U, Raaschou-Nielsen O, Ranzi A, Stephanou E, Sugiri D, Udvardy O, Vaskövi É, Weinmayr G, Brunekreef B, Hoek G. Spatial variation of PM_{2.5}, PM₁₀, PM_{2.5} absorbance and PM_{coarse} concentrations between and within 20 European study areas and the relationship with NO₂ - results of the ESCAPE project. *Atmos Environ*. 2012; 62:303–317.
- Gordon SB, Bruce NG, Grigg J, Hibberd PL, Kurmi OP, Lam KH, Mortimer K, Asante KP, Balakrishnan K, Balmes J, Bar-Zeev N, Bates MN, Breyse PN, Buist S, Chen Z, Havens D, Jack D, Jindal S, Kan H, Mehta S, Moschovis P, Naeher L, Patel A, Perez-Padilla R, Pope D, Rylance J, Semple S, Martin WJ. Respiratory risks from household air pollution in low and middle income countries. *The Lancet Respiratory Medicine*. 2014; 2:823–860. [PubMed: 25193349]

- Grahame TJ, Klemm R, Schlesinger RB. Public health and components of particulate matter: the changing assessment of black carbon. *J Air Waste Manage Assoc.* 2014; 64:620–660.
- He XZ, Chen W, Liu ZY, Chapman RS. An epidemiological study of lung cancer in Xuan Wei county, China: current progress. Case-control study on lung cancer and cooking fuel. *Environ Health Perspect.* 1991; 94:9–13. [PubMed: 1954946]
- Hosgood HD 3rd, Chapman R, Shen M, Blair A, Chen E, Zheng T, Lee KM, He X, Lan Q. Portable stove use is associated with lower lung cancer mortality risk in lifetime smoky coal users. *Br J Cancer.* 2008; 99:1934–1939. [PubMed: 19034286]
- Hosgood HD, Lan Q, Vermeulen R, Wei H, Reiss B, Coble J, Wei F, Jun X, Wu G, Rothman N. Combustion-derived nanoparticle exposure and household solid fuel use in Xuanwei and Fuyuan, China. *Int J Environ Health Res.* 2012; 22:571–581. [PubMed: 22639822]
- Hu W, Downward G, Reiss B, Rothman N, Xu J, Bassig B, Hosgood D, Zhang L, Wu G, Chapman R, Wei F, Lan Q, Vermeulen R. Personal and indoor fine particulate matter exposure from vented and unvented stoves in Xuanwei and Fuyuan county, China. *Environ Sci Technol.* 2014; 48:8456–64. [PubMed: 25003800]
- ISO. ISO 9835: ambient air — determination of a black smoke index. 1993
- Janssen NA, Hoek G, Simic-Lawson M, Fischer P, van Bree L, ten Brink H, Keuken M, Atkinson RW, Anderson HR, Brunekreef B, Cassee FR. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2.5. *Environ Health Perspect.* 2011; 119:1691–1699. [PubMed: 21810552]
- Jung KH, Patel MM, Moors K, Kinney PL, Chillrud SN, Whyatt R, Hoepner L, Garfinkel R, Yan B, Ross J, Camann D, Perera FP, Miller RL. Effects of heating season on residential indoor and outdoor polycyclic aromatic hydrocarbons, black carbon, and particulate matter in an urban birth cohort. *Atmos Environ* (1994). 2010; 44:4545–4552. [PubMed: 20938487]
- Kar A, Rehman IH, Burney J, Puppala SP, Suresh R, Singh L, Singh VK, Ahmed T, Ramanathan N, Ramanathan V. Real-time assessment of black carbon pollution in Indian households due to traditional and improved biomass cookstoves. *Environ Sci Technol.* 2012; 46:2993–3000. [PubMed: 22369148]
- Krämer U, Herder C, Sugiri D, Strassburger K, Schikowski T, Ranft U, Rathmann W. Traffic-related air pollution and incident type 2 diabetes: results from the SALIA cohort study. *Environ Health Perspect.* 2010; 118:1273–1279. [PubMed: 20504758]
- Lan Q, Chapman RS, Schreinemachers DM, Tian L, He X. Household stove improvement and risk of lung cancer in Xuanwei, China. *J Natl Cancer Inst.* 2002; 94:826–835. [PubMed: 12048270]
- Lanki T, Ahokas A, Alm S, Janssen NAH, Hoek G, De Hartog JJ, Brunekreef B, Pekkanen J. Determinants of personal and indoor PM2.5 and absorbance among elderly subjects with coronary heart disease. *Journal of Exposure Science and Environmental Epidemiology.* 2007; 17:124–133. [PubMed: 16519413]
- Lee KM, Chapman RS, Shen M, Lubin JH, Silverman DT, He X, Hosgood HD 3rd, Chen BE, Rajaraman P, Caporaso NE, Fraumeni JF Jr, Blair A, Lan Q. Differential effects of smoking on lung cancer mortality before and after household stove improvement in Xuanwei, China. *Br J Cancer.* 2010; 103:727–729. [PubMed: 20648014]
- Macintyre EA, Gehring U, Molter A, Fuertes E, Klumper C, Kramer U, Quass U, Hoffmann B, Gascon M, Brunekreef B, Koppelman GH, Beelen R, Hoek G, Birk M, de Jongste JC, Smit H, Cyrys J, Gruzdeva O, Korek M, Bergstrom A, Agius RM, de Vocht F, Simpson A, Porta D, Forastiere F, Badaloni C, Cesaroni G, Esplugues A, Fernandez-Somoano A, Lerxundi A, Sunyer J, Cirach M, Nieuwenhuijsen MJ, Pershagen G, Heinrich J. Air pollution and respiratory infections during early childhood: an analysis of 10 European birth cohorts within the ESCAPE Project. *Environ Health Perspect.* 2013; 122:107–113. [PubMed: 24149084]
- Martin WJ II, Hollingsworth JW, Ramanathan V. Chapter 13, Household air pollution from cookstoves: impacts on health and climate. 2014
- Mumford JL, He XZ, Chapman RS, Cao SR, Harris DB, Li XM, Xian YL, Jiang WZ, Xu CW, Chuang JC. Lung cancer and indoor air pollution in Xuan Wei, China. *Science.* 1987; 235:217–220. [PubMed: 3798109]

- Quincey P. A relationship between Black smoke index and black carbon concentration. *Atmos Environ*. 2007; 41:7964–7968.
- R Development Core Team. R: a language and environment for statistical computing. 2014
- Schmidt CW. Black carbon: the dark horse of climate change drivers. *Environ Health Perspect*. 2011; 119:A172–A175. [PubMed: 21459693]
- Shan M, Yang X, Ezzati M, Chaturvedi N, Coady E, Hughes A, Shi Y, Yang M, Zhang Y, Baumgartner J. A feasibility study of the association of exposure to biomass smoke with vascular function, inflammation, and cellular aging. *Environ Res*. 2014; 135:165–172. [PubMed: 25262090]
- Shen M, Chapman RS, Vermeulen R, Tian L, Zheng T, Chen BE, Engels EA, He X, Blair A, Lan Q. Coal use, stove improvement, and adult pneumonia mortality in Xuanwei, China: a retrospective cohort study. *Environ Health Perspect*. 2009; 117:261–266. [PubMed: 19270797]
- Tian L, Lan Q, Yang D, He X, Yu ITS, Hammond SK. Effect of chimneys on indoor air concentrations of PM10 and benzo[a]pyrene in Xuan Wei, China. *Atmos Environ*. 2009; 43:3352–3355.
- United Nations Environment Programme and World Meteorological Organization. Integrated assessment of black carbon and tropospheric ozone. 2011
- Van Vliet ED, Asante K, Jack DW, Kinney PL, Whyatt RM, Chillrud SN, Abokyi L, Zandoh C, Owusu-Agyei S. Personal exposures to fine particulate matter and black carbon in households cooking with biomass fuels in rural Ghana. *Environ Res*. 2013; 127:40–48. [PubMed: 24176411]
- World Health Organization. Fuel for Life: household energy and health. 2006

Practical Implications

This paper provides for the first time, an overview of the black carbon produced by domestic solid fuel combustion in an area of China with an exceptionally high lung cancer rate and highlights that there are multiple factors contributing to measurements. The finding that ventilated stoves, which have previously been found to reduce lung cancer rates, do not reduce black carbon illustrate the importance of reducing dependence upon solid fuels and moving populations up the energy ladder.

Figure 1. Map of Xuanwei and Fuyuan counties

Village location is indicated by designated numbers

Mines indicates are those reported by study participants and do not represent all mines present in the area

Personal and indoor PM_{2.5} absorbance measurements (in 10⁻⁵ m⁻¹) by fuel type and stove design**Table 1**

Fuel Type	Stove Design [‡]	Personal				Indoor			
		N(k)	AM	GM	GSD	N(k)	AM	GM	GSD
Smoky Coal	Overall	202(89)	13.8	12.0	1.7	209(87)	13.3	11.8	1.7
	Ventilated	108(48)	14.7	13.0	1.6	114(51)	13.6	12.2	1.7
	Unventilated	8(3)	10.5	10.0	1.4	8(3)	9.3	8.7	1.5
	Portable Stove	22(9)	8.9	7.3 [‡]	2.0	21(8)	10.1	7.7 [‡]	2.1
	Firepit	13(7)	14.4	13.7	1.4	13(6)	14.9	14.4	1.3
Smokeless Coal	Mixed	44(19)	15.3	13.5	1.8	45(15)	14.8	13.7	1.5
	Unknown	7(3)	7.8	7.2	1.6	8(4)	10.7	9.4	1.8
	Overall	46(20)	6.0	5.1[*]	1.7	45(18)	5.5	4.7[*]	1.8
	Ventilated	5(2)	7.6	6.1	2.0	5(2)	8.0	5.1 [*]	2.6
	Unventilated	17(6)	5.4	4.8	1.6	17(5)	5.6	5.1	1.6
“Other” Coal	Portable Stove	19(8)	6.1	5.1	1.9	18(7)	4.6	4.1	1.7
	Firepit	3(3)	3.8	3.7	1.3	3(3)	3.4	3.4	1.3
	Mixed	2(1)	10.0	9.9	1.2	2(1)	9.5	9.5	1.1
	Unknown	0	-	-	-	0	-	-	-
	Overall	37(20)	11.1	9.5	1.8	41(20)	9.6	8.1	1.9
Wood	Ventilated	13(6)	10.0	9.1	1.6	14(6)	10.2	9.0	1.7
	Unventilated	0	-	-	-	0	-	-	-
	Portable Stove	14(8)	11.5	9.2	2.2	14(8)	9.0	6.9	2.3
	Firepit	1(1)	9.8	9.8	-	1(1)	8.1	8.1	-
	Mixed	9(5)	12.4	10.8	1.7	12(5)	9.7	8.6	1.7
Wood	Unknown	0	-	-	-	0	-	-	-
	Overall	23(11)	17.4	16.3[*]	1.4	23(11)	15.5	15.0	1.3
	Ventilated	8(4)	14.6	13.5	1.5	8(4)	15.4	14.7	1.4
	Unventilated	0	-	-	-	0	-	-	-
	Portable Stove	6(2)	17.5	17.2	1.2	5(1)	14.8	14.5	1.3
Wood	Firepit	9(5)	19.8	18.6	1.4	9(5)	15.9	15.3	1.3

Fuel Type	Stove Design [‡]	Personal				Indoor			
		N(k)	AM	GM	GSD	N(k)	AM	GM	GSD
Plant	Mixed	0	-	-	-	0	-	-	-
	Unknown	0	-	-	-	1(1)	16.3	16.3	-
	Overall	13(9)	13.4	12.5	1.5	13(9)	13.0	12.6	1.3
	Ventilated	3(2)	11.0	10.9	1.2	3(2)	8.5	8.5	1.1
	Unventilated	3(1)	18.6	18.6	1.1	3(2)	15.0	15.0	1.0
	Portable Stove	2(1)	14.6	14.6	1.0	2(1)	14.2	13.9	1.3
	Firepit	4(4)	9.3	8.5	1.6	4(4)	13.6	13.2	1.3
	Mixed	1(1)	19.4	19.4	NA	1(1)	15.9	15.9	-
	Unknown	0	-	-	-	0	-	-	-
	Overall	93(48)	12.0	10.3	1.8	112(49)	12.3	10.1	2.0
“Other,” Fuel	Ventilated	17(8)	10.2	9.3	1.6	22(9)	10.6	9.1	1.8
	Unventilated	17(10)	13.5	11.4	2.0	26(11)	14.4	11.8	2.0
	Portable Stove	7(4)	9.5	8.5	1.7	7(4)	8.7	7.9	1.6
	Firepit	0	-	-	-	0	-	-	-
	Mixed	48(24)	12.9	11.1	1.8	53(23)	12.8	10.4	2.0
	Unknown	4(2)	7.2	6.4	1.8	4(2)	7.6	6.6	1.8
	Overall	103(52)	11.8	10.1	1.7	112(49)	11.8	10.1	1.8
	Ventilated	17(8)	10.2	9.3	1.6	22(9)	10.6	9.1	1.8
	Unventilated	17(10)	13.5	11.4	2.0	26(11)	14.4	11.8	2.0
	Portable Stove	7(4)	9.5	8.5	1.7	7(4)	8.7	7.9	1.6

[‡]Ventilated = stove with chimney, unventilated = stove without chimney, mixed = multiple stoves with different designs

^{*}Significant difference with smokey coal for same chimney strata (Tukey HSD test)

[‡]Significant difference with ventilated stove for that fuel type (Tukey HSD test)

N: Number of samples

k: Number of individuals subjects

Table 2

Personal and indoor PM_{2.5} absorbance measurements (in 10⁻⁵ m⁻¹) by smoky coal source

County	Smoky Coal Subtype	Mine Name	Personal				Indoor			
			N(k)	AM	GM	GSD	N(k)	AM	GM	GSD
<i>Xuanwei</i>	<i>Coking Coal</i>		117(47)	14.7	13.2 [*]	1.7	123(47)	14.7	13.2 [*]	1.7
		Azhi	32(13)	17.9	16.4	1.6	33(13)	18.3	17.1	1.5
		Baoshan	12(4)	13.8	12.9	1.5	12(4)	14.9	14.6	1.2
		Laibin	28(10)	11.6	10.7	1.6	31(10)	11.8	10.7	1.7
		Tangtang	31(14)	15.2	12.8	1.9	33(14)	14.2	11.9	1.9
		Yangchang	14(6)	13.3	12.9	1.3	14(6)	13.8	13.3	1.3
<hr/>										
<i>Fuyuan</i>	Coking Coal		85(39)	13.0	10.9 [†]	1.8	86(38)	11.5	10.3 [†]	1.7
			21(10)	10.5	9.4 [*]	1.7	25(10)	9.8	8.8 [*]	1.6
		Daping	9(4)	7.6	6.8	1.7	10(4)	6.6	6.0	1.5
		Enhong	7(4)	11.1	10.5	1.4	9(4)	10.8	10.4	1.4
		Haidan	5(2)	14.9	14.7	1.2	6(2)	13.5	13.1	1.4
	1/3 Coking coal		13(7)	11.1	10.4	1.4	12(7)	13.5	12.5	1.5
		Bagong	10(5)	12.2	11.6 [*]	1.4	9(5)	14.0	13.3	1.4
		Dahe	3(2)	7.3	7.1	1.3	3(2)	11.7	10.1	2.0
	Gas fat coal		40(20)	15.6	13.2	1.7	39(19)	12.9	12.1 [*]	1.5
		Housuo	38(19)	15.2	12.8	1.7	37(18)	12.4	11.7	1.4
	Meagre lean	Qingyun	2(1)	23.8	23.7	1.2	2(1)	21.5	21.3	1.2
		Gumu	4(2)	5.2	4.0	2.4	4(2)	3.4	3.0	1.8

^{*} Significant variation within designated coal subtype (ANOVA test)

[†] Significant variation between smoky coal subtypes within Fuyuan (ANOVA test)

N: Number of samples

k: Number of individual subjects

Table 3Determinants of personal and indoor PM_{2.5} absorbance measurements

Fuel Type	Personal			Indoor		
	Reference/Background Value (Intercept)*	Estimate	95% CI	GMR	Estimate	95% CI
			2.02			2.00
Smokeless Coal (FY & XW)	Ref	Ref	1	1	Ref	1
Coking Coal from north XW	0.45	0.15,0.74	1.56	0.3	0.03,0.58	1.35
Coking Coal from south XW	0.4	-0.11,0.89	1.49	0.23	-0.31,0.77	1.26
Coking Coal from FY	0.15	-0.16,0.47	1.17	-0.05	-0.34,0.24	0.95
1/3 Coking coal FY	0.33	-0.11,0.76	1.39	0.4	-0.06,0.84	1.49
Gas Fat coal FY	0.58	0.25,0.9	1.79	0.2	-0.14,0.54	1.23
Meagre Lean coal FY	-0.09	-0.67,0.5	0.92	-0.32	-0.9,0.26	0.73
Smoky Coal of uncertain type	0.35	-0.07,0.77	1.42	0.47	0.03,0.91	1.6
Multiple Coal Types	0.31	0.05,0.56	1.36	0.07	-0.17,0.31	1.07
Wood	0.8	0.48,1.11	2.22	0.51	0.21,0.81	1.67
Plant Products	0.5	0.15,0.84	1.64	0.3	-0.03,0.62	1.35
Multiple Fuel Types	0.39	0.16,0.62	1.48	0.21	0.0,0.42	1.23
Stove Design						
Ventilated	Ref	Ref	1	Ref	1	1
Unventilated	-0.11	-0.28,0.07	0.9	0.01	-0.16,0.18	1.01
Portable Stove	-0.17	-0.35,-0.002	0.84	-0.18	-0.35,-0.01	0.84
Mixed	-0.04	-0.17,0.1	0.96	-0.01	-0.14,0.12	0.99
Unknown	-0.49	-0.85,-0.12	0.61	-0.2	-0.5,0.1	0.82
Presence of Stairway						
Present	Ref	Ref	1	Ref	1	1
Absent	-0.13	-0.26,-0.01	0.88	-0.13	-0.26,-0.01	0.88
Unknown	-0.04	-0.84, 0.75	0.96	-0.24	-1,0.52	0.79
Season						
Autumn	Ref	Ref	1	Ref	1	1
Winter	0.28	0.13,0.43	1.32	0.17	0.02,0.32	1.19

Reference/Background Value (Intercept) *		Personal			Indoor		
Fuel Type	Estimate	95% CI	GMR	Estimate	95% CI	GMR	
Spring	-0.01	-0.14,0.12	0.99	-0.07	-0.2,0.05	0.93	
Summer	-0.16	-0.47,0.15	0.85	-0.26	-0.55,0.04	0.77	
Variance explained (%)							
Between Subjects/households		17				28	
Between Villages		66				47	

* Reference value represents log transformed absorbance value for the reference model entry (smokeless coal, burnt in a stove with a chimney in a room with a stair hole attached, during Autumn)

GMR: = geometric mean ratio = GM(estimate)/GM(reference)=exp(Estimate)

Example calculation: Personal BC value for “intercept” home = exp(2.02)=7.5

Personal BC value for home using coking coal from north Xuanwei = exp (2.02+0.45)=11.8, which corresponds to a GMR of 1.56

Table 4

Outdoor PM_{2.5} absorbance measurements (in 10⁻⁵ m⁻¹) by dominant fuel type[‡] used

Fuel Type [‡]	N(k)	AM	GM	GSD
Smoky Coal	38(14)	6.2	5.7	1.5
Smokeless Coal	10(3)	3.2	3.1 [*]	1.3
Other [‡]	18(7)	5.4	5.0 [*]	1.5

^{*} Significant difference when compared to Smoky coal (Tukey HSD test)

N: Number of samples

k: Number of individual villages

[‡] Refers to fuel used by > 50% of study enrollees per village

[‡] Refers to any other combination of fuels

Table 5Determinants of outdoor PM_{2.5} absorbance measurements

Reference/Background Value (Intercept)*		0.67		
	Estimate	95% CI	GMR	
Log-transformed indoor absorbance measurement	0.44	0.23,0.64	1.55	
Average outdoor humidity (%)	-0.01	-0.01,-0.001	0.99	
Season				
	Autumn	Ref	1	
	Winter	0.34	0.10,0.58	
	Spring/summer [†]	0.25	-0.01,0.50	
Variance explained%				
	Between Villages	52		

* Reference value represents log transformed absorbance value for the reference model entry (Indoor measurement(log transformed) and humidity measurements of zero, during Autumn)

GMR: = geometric mean ratio = GM(estimate)/GM(reference)=exp(Estimate)

[†] Merged due to few outdoor measurements in summer