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

Abstract 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. Measurements were taken over two 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. PM_{abs} measurements were higher in wood burning households (16.3×10^{-5}/m) than bituminous and anthracite coal households (12 and 5.1×10^{-5}/m, 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 ($r = 0.49$, $P < 0.01$). 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.

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Key words: Black carbon; Coal; China; Solid fuels; Household air pollution; Climate change; $PM_{2.5}$ absorbance.

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Practical Implications
This study 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.

Introduction

Xuanwei, and its neighboring 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 worldwide, are largely dependent upon the use of solid fuels (coal, wood, etc.) for their daily heating and cooking needs (World Health Organization, 2006), a practice which annually accounts for up to 4 million deaths

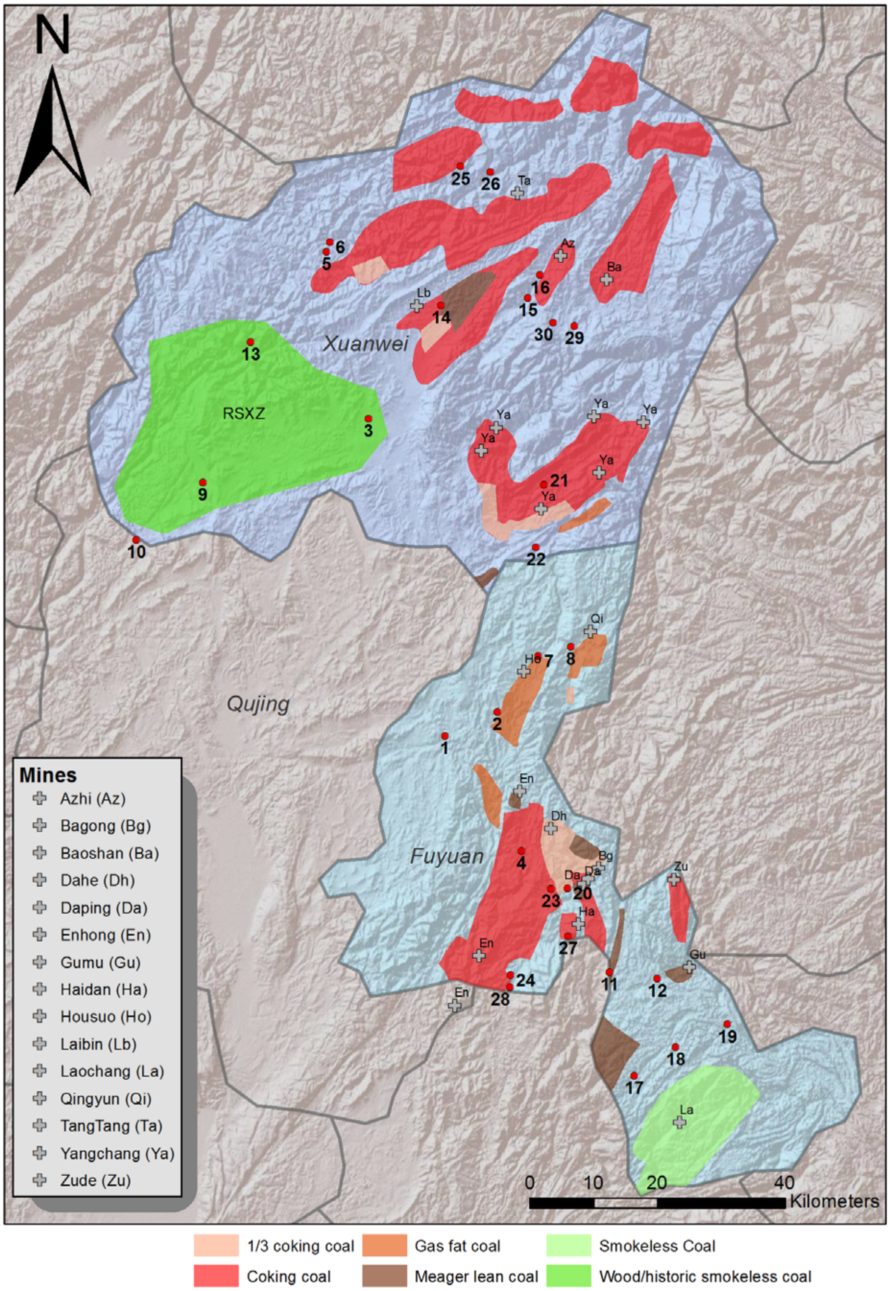

Fig. 1 Map of Xuanwei and Fuyuan counties. Village location is indicated by designated numbers. Mine indicates are those reported by study participants and do not represent all mines present in the area

worldwide (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 cookstoves 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 study 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.

Methods

Data for this study were 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 five households with a healthy non-smoking adult female resident were selected from each village. Household selection was directed toward 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 enrollment.

Homes were measured and sketched. Pertinent features (e.g., stoves, doors, windows, and stairways) were recorded. Data were collected during two 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 enrollees were re-visited and 15 new subjects were enrolled. During each collection period, measurements were taken during two consecutive 24-h 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 subtypes were classified according to the Chinese State Standard for coal classification [the subtypes are coking coal, 1/3 coking coal, gas fat coal, and meager 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).

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 cutoff of 2.5 μm (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 were not accepted for further analysis (approximately 2% of filters were considered not acceptable for use). Filters were changed after approximately 24 h 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 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 were established by accessing local records, collected by local health professionals that 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.

Sample analysis

Light reflectance was measured across five different spots on the filter (to compensate for variation across the filter) using an EEL 043 Smoke Stain Reflectometer. The average reflectance across the five points was transformed into an absorption value using the following formula (ISO, 1993).

$$a = \frac{A}{2V}\ln\left(\frac{R_0}{R_f}\right) \times 10^{-5}/\text{m} \qquad (1)$$

where a represents the absorption coefficient (in 10^{-5}/m), 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 (%).

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 eight personal measurements)]. The median relative percentage difference of these repeat measurements was 7% [interquartile range (IQR) 3.4–13.5%].

Comparison to other components of air pollution

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

Statistical analysis

Normal probability plots indicated that measured absorbance values tended toward 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.

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:

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

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 <0.05 was considered to indicate statistical significance.

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 meager 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.

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) 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 (16.3×10^{-5}/m) and indoor (15.0×10^{-5}/m) measurements, and for personal measurements, they were significantly higher than households burning smoky coal (12×10^{-5}/m, $P < 0.05$). Households exclusively burning smoky coal had significantly higher PM_{abs} than households burning smokeless coal for both personal (12×10^{-5}/m vs. 5.1×10^{-5}/m, $P < 0.05$) and indoor (11.8×10^{-5}/m vs. 4.7×10^{-5}/m, $P < 0.05$) 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 7.7×10^{-5}/m vs. 12.0 and 12.2×10^{-5}/m, 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 (23.7×10^{-5}/m and 21.3×10^{-5}/m, respectively) while the coking coal mine 'Daping' also located in Fuyuan had the lowest (6.8×10^{-5}/m and 6.0×10^{-5}/m). ANOVA testing revealed significant variation within coking coals sourced from within Xuanwei (range for personal measurements 10.7×10^{-5}/m to 16.4×10^{-5}/m, $P < 0.05$), within the smoky coal subtypes produced in Fuyuan (range 5.2×10^{-5}/m for meager lean coal to 13.2×10^{-5}/m for gas fat coal) and within the Fuyuan coking coals (6.8×10^{-5}/m to 14.7×10^{-5}/m), 1/3 coking coals (7.1×10^{-5}/m to 11.6×10^{-5}/m, personal measurements only), and the gas fat coal subtypes (11.7×10^{-5}/m to 21.3×10^{-5}/m, indoor measurements only).

Mixed effect modeling. 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 = GM(estimate)/GM(reference) = exp(β)) 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

Table 1 Personal and indoor $PM_{2.5}$ absorbance measurements (in 10^{-5}/m) by fuel type and stove design

		Personal				Indoor			
Fuel type	Stove design[a]	*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[c]	2.0	21 (8)	10.1	7.7[c]	2.1
	Firepit	13 (7)	14.4	13.7	1.4	13 (6)	14.9	14.4	1.3
	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
Smokeless coal	Overall	**46 (20)**	**6.0**	**5.1[b]**	**1.7**	**45 (18)**	**5.5**	**4.7[b]**	**1.8**
	Ventilated	5 (2)	7.6	6.1	2.0	5 (2)	8.0	5.1[b]	2.6
	Unventilated	17 (6)	5.4	4.8	1.6	17 (5)	5.6	5.1	1.6
	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	–	–	–
'Other' coal	Overall	**37 (20)**	**11.1**	**9.5**	**1.8**	**41 (20)**	**9.6**	**8.1**	**1.9**
	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
	Unknown	0	–	–	–	0	–	–	–
Wood	Overall	**23 (11)**	**17.4**	**16.3[b]**	**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
	Firepit	9 (5)	19.8	18.6	1.4	9 (5)	15.9	15.3	1.3
	Mixed	0	–	–	–	0	–	–	–
	Unknown	0	–	–	–	1 (1)	16.3	16.3	–
Plant	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	–	–	–
'Other' fuel	Overall	**93 (48)**	**12.0**	**10.3**	**1.8**	**112 (49)**	**12.3**	**10.1**	**2.0**
	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

N, number of samples; *k*, number of individuals subjects; AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation.
[a]Ventilated, stove with chimney; unventilated, stove without chimney; mixed, multiple stoves with different designs.
[b]Significant difference with smoky coal for same chimney strata (Tukey HSD test).
[c]Significant difference with ventilated stove for that fuel type (Tukey HSD test).
Bold represent overall values for individual fuel types.

meager 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).

Comparisons to other air measurements. Previous research on this population has presented personal and indoor particulate mass ($PM_{2.5}$) measurements (Hu et al., 2014). In brief, this study found that, similarly to the current study, $PM_{2.5}$ measurements were higher among smoky coal users (148 $\mu g/m^3$) than smokeless (115 $\mu g/m^3$) but lower than wood users (289 $\mu g/m^3$). Unlike the current study, $PM_{2.5}$ measurements were lower among users of ventilated stoves than users of

Table 2 Personal and indoor $PM_{2.5}$ absorbance measurements (in 10^{-5}/m) by smoky coal source

County	Smoky coal subtype	Mine name	Personal				Indoor			
			N(k)	AM	GM	GSD	N(k)	AM	GM	GSD
Xuanwei	*Coking Coal*		*117 (47)*	*14.7*	*13.2* [a]	*1.7*	*123 (47)*	*14.7*	*13.2* [a]	*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
Fuyuan			*85 (39)*	*13.0*	*10.9* [b]	*1.8*	*86 (38)*	*11.5*	*10.3* [b]	*1.7*
	Coking coal		21 (10)	10.5	9.4[a]	1.7	25 (10)	9.8	8.8[a]	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[a]	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[a]	1.5
		Housuo	38 (19)	15.2	12.8	1.7	37 (18)	12.4	11.7	1.4
		Qingyun	2 (1)	23.8	23.7	1.2	2 (1)	21.5	21.3	1.2
	Meager lean	Gumu	4 (2)	5.2	4.0	2.4	4 (2)	3.4	3.0	1.8

N, number of samples; *k*, number of individual subjects; AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation.
[a]Significant variation within designated coal subtype (ANOVA test).
[b]Significant variation between smoky coal subtypes within Fuyuan (ANOVA test).
Italics represent overall values for each county.

firepits (values for smoky coal users: 150 μg/m^3 for vented stoves and 307 μg/m^3 for firepits, $P < 0.05$). The ratio of PM_{abs} to $PM_{2.5}$ 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 $PM_{2.5}$ measurements showed a strong degree of correlation with each other (Spearman r: 0.68, $P < 0.05$).

Measurements of 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 $PM_{2.5}$ 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 PM_{abs}:BaP ratio for smoky coal burnt in a firepit was 1:11 while for smoky coal in a vented stove it was 1:3.

Village-based outdoor measurements

Village-based outdoor measurements correlated moderately well with both the average indoor and personal measurements of study enrollees 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: 5.7×10^{-5}/m) and lowest among smokeless coal-using villages (GM: 3.1×10^{-5}/m, 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

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

	Personal			Indoor		
Reference/background value (intercept)[a]		2.02			2.00	
Fuel type	Estimate	95% CI	GMR	Estimate	95% CI	GMR
Smokeless coal (FY & XW)	Ref		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
Meager 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, 0.42	1.23
Stove design						
Ventilated	Ref		1	Ref		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		1	Ref		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		1	Ref		1
Winter	0.28	0.13, 0.43	1.32	0.17	0.02, 0.32	1.19
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	

[a]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) by dominant fuel type[b] used

Fuel type[b]	*N*(*k*)	AM	GM	GSD
Smoky coal	38 (14)	6.2	5.7	1.5
Smokeless coal	10 (3)	3.2	3.1[a]	1.3
Other[c]	18 (7)	5.4	5.0[a]	1.5

N, number of samples; *k*, number of individual villages; AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation.
[a]Significant difference when compared to Smoky coal (Tukey HSD test).
[b]Refers to fuel used by >50% of study enrollees per village.
[c]Refers to any other combination of fuels.

Table 5 Determinants of outdoor $PM_{2.5}$ absorbance measurements

Reference/background value (Intercept)[a]	Estimate	0.67 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	1.41
Spring/summer[b]	0.25	−0.010.50	1.28
Variance explained%			
Between Villages		52	

[a]Reference value represents log-transformed absorbance value for the reference model entry (indoor measurement (log-transformed) and humidity measurements of zero, during Autumn).
[b]Merged due to few outdoor measurements in summer.
GMR = geometric mean ratio = GM (estimate)/GM (reference) = exp(Estimate).

number of external pollution sources did not contribute to the final model.

Discussion

Black carbon emissions from the domestic combustion of solid fuels have 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

$PM_{2.5}$ 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).

Enrollees burning wood were found to have the highest personal PM_{abs} levels (16.3×10^{-5}/m) followed by plant and smoky coal (12.5 and 12.0×10^{-5}/m, 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 1.26×10^{-5}/m 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) 0.97×10^{-5}/m (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 (6×10^{-5}/m) are frequently in excess of those by a European urban roadside. For example, several large multicentre studies exploring air pollution throughout Europe have reported average absorbance concentrations of $<2 \times 10^{-5}$/m (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 BC, using a conversion to black smoke as an intermediate step (Quincey, 2007). This conversion results in an average personal BC 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 BC 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 cookstove 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 nonmalignant 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 unforced 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 airflow 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 underestimated. To assess any potential impact of this under-estimation on the main findings of the current study, two separate sensitivity analyses were conducted. First, we analyzed 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 study. 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 study (specifically the roles that fuel and stove types play in measurements) are robust.

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 BC, 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 are 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 HAP is warranted.

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Competing financial interests declaration

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

Supporting Information

Additional Supporting Information may be found in the online version of this article:

Table S1. Variables considered for inclusion in linear mixed models.

Table S2. Determinants of the differences between personal and indoor measurements.

Figure S1. Scatter plot showing relationship between average personal (left) and average indoor (right) measurements with village based measurements (all measurements in 10^{-5}/m).

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