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Indoor concentrations of nitrogen dioxide and sulfur dioxide from burning solid fuels for cooking and heating in Yunnan Province, China

Abstract The Chinese national pollution census has indicated that the domestic burning of solid fuels is an important contributor to nitrogen dioxide (NO_2) and sulfur dioxide (SO_2) emissions in China. To characterize indoor NO_2 and SO_2 air concentrations in relation to solid fuel use and stove ventilation in the rural counties of Xuanwei and Fuyuan, in Yunnan Province, China, which have among the highest lung cancer rates in the nation, a total of 163 participants in 30 selected villages were enrolled. Indoor 24-h NO_2 and SO_2 samples were collected in each household over two consecutive days. Compared to smoky coal, smokeless coal use was associated with higher NO_2 concentrations [geometric mean (GM) = 132 $\mu g/m^3$ for smokeless coal and 111 $\mu g/m^3$ for smoky coal, $P = 0.065$] and SO_2 [limit of detection = 24 $\mu g/m^3$; percentage detected (% Detect) = 86% for smokeless coal and 40% for smoky coal, $P < 0.001$]. Among smoky coal users, significant variation of NO_2 and SO_2 air concentrations was observed across different stove designs and smoky coal sources in both counties. Model construction indicated that the measurements of both pollutants were influenced by stove design. This exposure assessment study has identified high levels of NO_2 and SO_2 as a result of burning solid fuels for cooking and heating.

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Key words: Coal; Household air pollution; Indoor; Nitrogen dioxide; Sulfur dioxide; Ventilation.

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Practical Implications
Smokeless (anthracite) coal is generally regarded as a safer alternative to smoky (bituminous) coal, although studies have shown the former to be linked to increased risk of respiratory diseases. In this study, we reported that smokeless coal was associated with higher levels of household nitrogen dioxide (NO_2) and a higher percentage of detectable sulfur dioxide (SO_2) than smoky coal in the rural counties of Xuanwei and Fuyuan, in Yunnan Province, China, which use a variety of solid fuels including smoky coal, smokeless coal, and wood. Our findings support the WHO guidelines to reduce reliance on household coal use and prioritize use of clean fuels such as gas or electricity.

Introduction

The domestic burning of coal or biomass for cooking and heating is practiced by approximately three billion people worldwide, mostly with poorly ventilated stoves in low- and middle-income countries, including China. Coal combustion has been reported to emit large amounts of SO_2 and NO_2 and contribute dominantly to elevated indoor NO_2 and SO_2 levels that exceed China's indoor air quality standards (Zhang and Smith, 2007). The first national pollution census in China in 2010 reported that 8.60% of total SO_2 emissions and 3.24% of total NO_x emissions came from domestic sources due to coal combustion for cooking and heating (China Internet Information Center, 2010). Beyond this census, there is limited exposure data available on indoor NO_2 and SO_2 air concentrations in China, especially in rural areas using solid fuels for heating and cooking. Most people spend around 90% of their time indoors and the majority of their indoor time at home, and hence, they may be chronically exposed to the air pollutants produced from household solid fuel use (Leech et al., 2002; Zhao et al., 2009).

Both NO_2 and SO_2 have been shown to be associated with chronic diseases of the respiratory system especially asthma, chronic obstructive pulmonary disease (COPD), and cardiovascular diseases (Anderson et al., 1997; Ko et al., 2007; Atkinson et al., 2013). A meta-analysis of the long-term adverse health effects of NO_2 and SO_2 in Chinese populations found increased risks of respiratory mortality for both NO_2 and SO_2 (Lai et al., 2013). Additionally, each 10-ppb increase in SO_2 from indoor heating sources has been shown to increase wheezing and chest tightness among non-smoking women in the United States (Triche et al., 2005). It is also possible that indoor NO_2 and SO_2 levels may affect outdoor air quality and contribute to climate change (U.S. Environmental Protection Agency, 2010).

In the neighboring counties of Xuanwei and Fuyuan, the negative health effects of solid fuel use for heating and cooking are well illustrated. In these counties, the combustion of locally sourced 'smoky' coal (a bituminous coal) is associated with increased malignant and non-malignant lung diseases and notably one of the highest lung cancer rates in the nation (Mumford et al., 1987; Shen et al., 2009). Previous studies on this population have demonstrated significant variation of polycyclic aromatic hydrocarbons (PAHs) (Downward et al., 2014b) and $PM_{2.5}$ levels by fuel type, coal source, and stove design (Hu et al., 2014) and also reported heterogeneity in coal composition between and within coal types (Downward et al., 2014a). Specifically, these studies reported higher PAHs, and $PM_{2.5}$ air concentrations in households using smoky coal than smokeless coal (Downward et al., 2014b; Hu et al., 2014). Measurements were also higher for fuels burnt in unventilated firepits than in ventilated stoves. Smoky coal also contained higher levels of carbon, volatile organic matter, and quartz, but lower levels of trace and major elements (Al, Ti, Na, K, P, Cr, Ba, and Zr) compared to smokeless coal (Downward et al., 2014a).

There is limited literature evaluating indoor NO_2 and SO_2 levels between different coal and stove types, especially in this region. Here, we report our assessment of indoor NO_2 and SO_2 and the comparison of these measurements across different fuel types, coal regions, and stove designs.

Materials and methods

Study population

The data collected for this study is part of a large cross-sectional molecular epidemiology study aimed at categorizing solid fuel emissions from throughout Xuanwei and Fuyuan before ultimately matching those emissions with biological effect markers and disease risk in a case–control study of never-smoking females. The current study population consists of 163 households and their female heads from 30 villages in Xuanwei and Fuyuan counties of Yunnan Province in China. Villages and households were selected to have a representative coverage of the major geographical regions, fuel types, and stove designs in Xuanwei and Fuyuan. Furthermore, selection was directed toward reflecting the population in the case–control study; therefore, households which were at least 10 years old and had not had any stove alterations undertaken within the past 5 years were preferentially selected.

The study was carried out in two phases: Phase I was carried out from August 2008 to February 2009 with all 30 villages visited and 148 participants enrolled; Phase II was carried out between March and June of 2009 with 16 of the villages revisited, 15 newly enrolled participants, and 53 of the participants from the first phase revisited. Further details of this study population can be found in an earlier report of indoor and outdoor fine particulate matter ($PM_{2.5}$) (Hu et al., 2014). All study participants provided written informed consent prior to participation, and the study protocol was approved by institutional review boards of the China National Environmental Monitoring Center and the U.S. National Cancer Institute.

Questionnaires and household interviews

Details of the household interviews were described previously (Hu et al., 2014). Briefly, two trained interviewers conducted interviews with each household and administered a short activity questionnaire, which included information on cooking activities, heating practices, fuel use, and hours spent indoors. In addition, homes sketched with pertinent details (e.g. doors and windows) were being recorded. Detailed information on fuel type ('smoky' or 'smokeless' coal, wood, plant products, manufactured coal briquettes, combinations of briquettes, smoky, and smokeless coal ('mixed' coal), and combinations of wood, plant materials and coal ('mixed' fuel)) was reported. Stove design was also recorded at the time of interview. The major stove designs included vented stoves (stoves connected to a chimney), unvented stoves (stoves without chimneys), firepits, and portable stoves (a stove intended to be lit outdoors and then carried indoors for use), all manufactured locally. When households used multiple differing stove designs, we referred to these as 'mixed ventilation'. Smoky and smokeless coal types were confirmed by geochemical analysis of whole coal samples (Downward et al., 2014a) and were further classified by coal mines of origin based on coal mine location (coal source) and subtypes (smokeless, coking, 1/3 coking, gas fat, and meager lean coals) based on State Standard of China Coal Classification (GB5751-86) (Chen, 2000).

Exposure assessment of NO_2 and SO_2

Indoor measurements of NO_2 and SO_2 were collected using Ogawa passive badges (Ogawa & Co., Pompano Beach, FL, USA). Samplers were placed in the main cooking area of the home, approximately 2 m from the nearest wall and stove (as allowed by the available space) and remained for approximately 24-h (median measurement period 23-h). After sampling, filters were individually packaged into 5-ml sampling vials and stored at −80°C until analysis. For quantification of NO_2 and SO_2, filters were placed in glass vials with 5 ml of ultra-pure water and 0.1 ml of 3.5% H_2O_2 and shaken for 10 s every 2 min for a total period of 10 min. Each filter was analyzed for both NO_2 and SO_2. Extracts were analyzed using ion chromatography (IC) on a Dionex. For each run, internal laboratory standards were used to derive a standard curve for NO_2 and SO_2. NO_2 and SO_2 concentrations (in mg/l) were subsequently converted into parts per billion (ppb) as a function of sampling time and temperature conditions during the sampling period before conversion to micrograms of gaseous pollutant per cubic meter of ambient air ($\mu g/m^3$), assuming an ambient pressure of 1 atmosphere and a temperature of 25°C, for comparisons with national standards and other literature. Samples with readings above the standard curve were rerun at a dilution of 1:10. The limit of detection (LOD) for this methodology was calculated as a function of three times the standard deviation of field blanks [0.2 mg/l for NO_2 (approximately 75 $\mu g/m^3$) and 0.06 mg/l for SO_2 (approximately 24 $\mu g/m^3$)].

A total of 405 measurements were collected, of which 215 (53.1%) SO_2 measurements and 59 (14.6%) NO_2 measurements were below the LOD. Non-detects among the NO_2 samples were mathematically imputed using a multiple imputation procedure (Lubin et al., 2004). There were insufficient detected SO_2 samples for mathematical imputation. We therefore treated the SO_2 data both continuously and dichotomously as detected or non-detected. Approximately 10% of the households were sampled twice (1 year apart) for quality control. The overall coefficient of variation was 26.6% for NO_2, and the intraclass correlation coefficient of duplicate samples was 67.9% for NO_2. The percentage agreement in detect vs. non-detect between duplicate samples for SO_2 was 90.0%.

Statistical analysis

The concentrations of NO_2 approximated a log-normal distribution and values were natural logarithm-transformed before use in statistical tests. For NO_2, arithmetic mean, geometric mean (GM), and geometric standard deviation (GSD) were calculated for each fuel type and stove design individually, as well as each fuel type–stove design combination. The Tukey's honest significant difference test was used to compare the NO_2 levels between each fuel type with smoky coal, and also between each stove design with ventilated stove within each fuel type. The ANOVA test was then used to test for NO_2 differences among different stove designs within each fuel type. Due to the large proportion of undetectable values of SO_2 measurements (53.1%), detection rate (%Detect) was calculated for each fuel type and stove design individually, as well as each fuel type–stove design combination. Median levels were also calculated after replacing non-detects by $LOD/\sqrt{2}$. SO_2 was analyzed as a dichotomous variable (detected or non-detected) using logistic regression and Fisher's exact tests. Mixed effects linear models were used to identify determinants of NO_2, and mixed effects logistic models were used for SO_2. Subjects and villages were treated as random factors (random intercepts for each subject), whereas stove design, fuel type/source, seasonality, and other individual factors (Table S1) were considered for inclusion as fixed effects. The final model consisted of variables that resulted in the lowest Akaike information criterion. The mixed effects linear model can be represented with the following formula:

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

where y_{ijf} represents the measurements for village i, household j on day f. μ represents the intercept (i.e. the 'background' level). β_1 through β_n represents 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 household j, living in village i. ε_{ijf} represents the error for village i, household j on day f.

All statistical analyses were conducted using R, version 3.0.3 (R Core Team, 2014), and SAS, version 9.3 (SAS Institute Inc., Cary, NC, USA). All statistical tests were conducted as two-sided, and a *P*-value of <0.05 was considered to be statistically significant.

Results

Characteristics of the study population have been reported (Table S2) (Hu et al., 2014). Smoky coal ($n = 99$, 45.8%) and ventilated stoves ($n = 75$, 34.7%) were most commonly used. The mean age of the study population was 56 years (SD = 14.4 years), and the median time of stove use was 5.1 h per day. The overall GM concentration level of indoor NO_2 was 115 $\mu g/m^3$ (GSD = 1.45). SO_2 had an overall detection rate of 47%.

Indoor NO_2

Overall, measurements from smokeless coal using households had higher levels of NO_2 than smoky coal households across different stove designs (GM = 132 $\mu g/m^3$ for smokeless coal and 111 $\mu g/m^3$ for smoky coal, $P = 0.065$) (Table 1). There was significant variation of NO_2 levels across different stove designs for smoky coal users ($P < 0.001$) and mixed coal users ($P = 0.036$). Among smoky coal users, the use of firepits resulted in significantly higher levels of NO_2, compared to those with ventilated stoves (GM = 132 $\mu g/m^3$ for firepits and 102 $\mu g/m^3$ for ventilated stove, $P = 0.048$). Mixed ventilation had significantly higher measurements of NO_2, compared to ventilated stoves among smoky coal users (GM = 137 $\mu g/m^3$ for mixed stoves and 102 $\mu g/m^3$ for ventilated stoves, $P < 0.001$) and mixed coal users (median = 133 $\mu g/m^3$ for mixed stoves and 90 $\mu g/m^3$ for ventilated stoves, $P = 0.052$).

We observed heterogeneity in NO_2 concentrations between the different smoky coal sources for the single smoky coal subtype used in Xuanwei (coking coal, range of GM = 93–138 $\mu g/m^3$, $P < 0.001$). In Fuyuan, we observed heterogeneity of NO_2 concentrations across the smoky coal subtypes available within Fuyuan ($P < 0.001$) and within the coking coal subtype in Fuyuan (range of GM = 86–139 $\mu g/m^3$, $P = 0.0044$) (Table 2). The construction of a linear mixed model indicated that stove ventilation, season, and the average number of hours of burning fuel were found to be significant determinants of indoor NO_2 concentrations. Effect estimates (β), 95% confidence intervals (CIs), and geometric mean ratios (GMRs) (GMR = GM[estimate]/GM[reference] = exp[β]) are available in Table 3. The constructed model explains 16.7% and 43.0% of the variance between households and villages, respectively. It indicates that homes burning fuel in firepits had the highest NO_2 levels (GMR = 1.21, 95% CI = 1.04–1.40) and that measurements taken during spring/summer were the highest (GMR = 1.14, 95% CI = 1.04–1.24). The number of hours spent using stoves also positively predicted NO_2 levels. Every hour of stove use would result in a GMR increase of 1.01 (95% CI = 1.01–1.02).

Indoor SO_2

The proportion of smokeless coal using households where SO_2 measurements were above the LOD (86%), was significantly higher than smoky coal using households (40%, $P < 0.05$) (Table 1). The median concentration was also higher in smokeless coal (907 $\mu g/m^3$) compared to smoky coal use (<LOD). Mixed coal use and mixed fuel use also resulted in significantly higher detection rates of SO_2 than smoky coal (51% and 53% respectively). There was not only significant variation in detection rates across different stove designs among smoky coal ($P = 0.0024$) and smokeless coal ($P = 0.047$) users, but also among mixed coal ($P = 0.0012$) and mixed fuel ($P = 0.010$) users. Among smoky coal users, the use of firepits resulted in significantly higher SO_2 detection rates than ventilated stoves (73% [median = 234 $\mu g/m^3$] and 31% [median < LOD], respectively, $P = 0.0049$). Households using unventilated stoves (69% [median = 74], $P = 0.012$) and mixed stoves (57% [median = 34], $P = 0.023$) also had higher detection rates of SO_2 than households using ventilated stoves (25% [median < LOD]) for mixed fuel users.

Overall, we observed heterogeneity of SO_2 detection rates in Fuyuan (range of %Detect = 32–100%, $P = 0.0019$) (Table 2). The detection rates of SO_2 emissions from coking coal are significantly higher in Fuyuan (60%) compared to Xuanwei (29%) ($P = 0.0051$). Construction of a mixed effects logistic model indicated that stove ventilation and fuel type were significant predictors of detectable indoor SO_2 measurements (Table 3). The constructed model explains 24.4% and 36.7% of the variance between households and villages, respectively. It indicates that homes burning fuel in unventilated stoves and firepits had the highest rate of SO_2 detection. For example, the odds ratio of detectable SO_2 measurements among homes using unventilated stoves was 9.63 when compared to homes using ventilated stoves ($P = 0.023$).

Discussion

In this paper, we quantified indoor NO_2 and SO_2 air concentrations in two rural Chinese regions with a high-incidence of malignant and non-malignant lung diseases and showed that measurements differed by fuel type, stove design, and smoky coal source. Overall, both indoor NO_2 and SO_2 concentrations were found to be higher among smokeless coal users; however, there was also significant heterogeneity by stove design and smoky coal source. For instance, the use of mixed stoves (most probably due to the contribution of

Table 1 Indoor NO_2 and SO_2 air concentrations, by different fuel types and stove ventilation

	NO_2 ($\mu g/m^3$)								SO_2 ($\mu g/m^3$)							
	N	AM	GM	GSD	P^a	P^b	P^c	P^d	*N*	Median^e	IQR^e	%Detect	P^f	P^g	P^h	P^i
Smoky coal	191	118	111	1.42	Ref	–	–	–	191	<LOD	37	40	Ref	–	–	–
Ventilated stove	105	107	102	1.37	–	Ref	**<0.001**	0.077	105	<LOD	19	31	–	Ref	**0.0024**	0.22
Unventilated stove[j]	6	114	113	1.12	–	0.98	–	0.62	6	47	40	67	–	0.97	–	**0.0092**
Portable stove	19	130	118	1.57	–	0.48	–	0.47	19	35	270	63	–	0.12	–	**0.00048**
Firepit	15	135	132	1.21	–	**0.048**	–	0.97	15	234	838	73	–	**0.0049**	–	0.052
Mixed ventilation stove[k]	36	146	137	1.45	–	**<0.001**	–	0.61	36	<LOD	32	44	–	0.19	–	**0.0061**
Smokeless coal	42	145	132	1.56	0.065	–	–	–	42	907	1618	86	**<0.001**	–	–	–
Ventilated stove	5	144	135	1.52	–	Ref	0.99	–	5	2108	3240	80	–	Ref	**0.047**	–
Unventilated stove	15	146	134	1.52	–	1.00	–	–	15	224	888	93	–	0.41	–	–
Portable stove	17	142	127	1.65	–	1.00	–	–	17	896	1495	88	–	0.39	–	–
Firepit	3	161	139	1.90	–	1.00	–	–	3	1523	225	100	–	0.86	–	–
Mixed ventilation stove	2	147	147	1.08	–	1.00	–	–	2	<LOD	0	0	–	–	–	–
Mixed coal[l]	41	115	109	1.43	0.99	–	–	–	41	26	71	51	0.18	–	–	–
Ventilated stove	13	96	90	1.48	–	Ref	**0.036**	–	13	<LOD	0	23	–	Ref	**0.0012**	–
Unventilated stove	0	–	–	–	–	–	–	–	0	–	–	–	–	–	–	–
Portable stove	18	115	110	1.36	–	0.33	–	–	18	<LOD	49	44	–	0.29	–	–
Firepit	2	151	151	1.09	–	0.18	–	–	2	706	126	100	–	0.16	–	–
Mixed ventilation stove	8	138	133	1.32	–	**0.052**	–	–	8	163	174	100	–	0.99	–	–
Wood	24	115	110	1.36	0.99	–	–	–	24	<LOD	10	29	0.32	–	–	–
Ventilated stove	8	108	104	1.38	–	Ref	0.17	–	8	<LOD	12	38	–	Ref	0.40	–
Unventilated stove	0	–	–	–	–	–	–	–	0	–	–	–	–	–	–	–
Portable stove	5	92	90	1.24	–	0.85	–	–	5	<LOD	0	0	–	0.41	–	–
Firepit	10	134	128	1.35	–	0.43	–	–	10	<LOD	11	40	–	1.00	–	–
Mixed ventilation stove	1	93	93	–	–	0.99	–	–	1	<LOD	0	0	–	–	–	–
Plant materials[m]	14	127	119	1.43	0.98	–	–	–	14	<LOD	0	7	–	–	–	–
Ventilated stove	3	80	80	1.07	–	Ref	0.19	–	3	<LOD	0	0	–	Ref	1.00	–
Unventilated stove	3	116	114	1.27	–	0.66	–	–	3	<LOD	0	0	–	–	–	–
Portable stove	2	164	164	1.06	–	0.17	–	–	2	<LOD	0	0	–	–	–	–
Firepit	5	148	135	1.55	–	0.23	–	–	5	<LOD	0	20	–	1.00	–	–
Mixed ventilation stove	1	126	126	–	–	0.73	–	–	1	<LOD	0	0	–	–	–	–
Mixed fuel[n]	93	130	121	1.47	0.44	–	–	–	93	26	500	53	**0.041**	–	–	–
Ventilated stove	20	124	117	1.45	–	Ref	0.19	–	20	<LOD	3	25	–	Ref	**0.010**	–
Unventilated stove	16	149	138	1.47	–	0.68	–	–	16	74	719	69	–	**0.012**	–	–
Portable stove	3	107	107	1.12	–	1.00	–	–	3	284	199	100	–	0.99	–	–
Firepit	0	–	–		–	–	–	–	0	–	–	–	–	–	–	–
Mixed ventilation stove	53	129	121	1.47	–	1.00	–	–	53	34	799	57	–	**0.023**	–	–

AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation; %Detect, detection rate; <LOD, values below the limit of detection (LOD).
Significant values are indicated in bold.
[a] *P*-values from Tukey test, comparing each fuel type with smoky coal, using log-transformed NO_2 values.
[b] *P*-values from Tukey test, comparing each stove type with ventilated stove in same fuel type, using log-transformed NO_2 values.
[c] *P*-values from ANOVA test, comparing between stove types within each fuel type, using log-transformed NO_2 values.
[d] *P*-values from ANOVA test, comparing between fuel types within each stove type, using log-transformed NO_2 values.
[e] Non-detects were replaced by LOD/square root(2). IQR was calculated by taking the difference between the 3rd and 1st quartile.
[f] *P*-values from logistic regression, comparing detected/non-detected SO_2 of each fuel type with smoky coal.
[g] *P*-values from logistic regression, comparing detected/non-detected SO_2 of each stove type with ventilated stove in same fuel type.
[h] *P*-values from Fisher's exact test, comparing detected/non-detected SO_2 between stove types within each fuel type.
[i] *P*-values from Fisher's exact test, comparing detected/non-detected SO_2 between fuel types within each stove type.
[j] Includes high and/or low stoves without any chimney.
[k] Mixed ventilation refers to a mixture of different stove designs.
[l] Includes the use of smoky, smokeless coal and prepared coal briquettes.
[m] Includes the use of wood, tobacco stem, and corncob.
[n] Includes the use of wood, plant materials, and coal.

unvented stoves) and firepits was associated with significantly higher NO_2 than ventilated stoves among smoky coal users. Indoor NO_2 and SO_2 levels also showed significant variation by the coal mine region where the coal originated (coal source).

In a study of indoor NO_2 concentrations and the risk of childhood acute leukemia in Shanghai, where the majority of the households (more than 95%) used natural gas or liquefied petroleum gas for fuel, the maximum indoor 24-h NO_2 concentration in the child's bedroom was 104 $\mu g/m^3$, which is less than the mean NO_2 of smoky and smokeless coal households in our study, regardless of stove type (Gao et al., 2014). A survey conducted in four cities in China (Chengde,

Table 2 Indoor NO_2 and SO_2 air concentrations from smoky coal burning homes from Xuanwei and Fuyuan, by coal source

				NO_2 (μg/m^3)							SO_2 (μg/m^3)						
County	Coal type	Coal subtype	Coal source	N[a]	AM	GM	GSD	P[b]	P[c]	P[d]	N[a]	Median[e]	IQR[e]	%Detect	P[f]	P[g]	P[h]
Xuanwei	Smoky	Overall		110	121	114	1.42	–	–	–	110	<LOD	12	29	–	–	–
		Coking Coal						–	**<0.001**	0.68					–	0.073	**0.0051**
			Azhi	30	148	138	1.47		–		30	<LOD	16	33	–	–	
			Baoshan	12	134	130	1.26	–	–		12	<LOD	87	42	–	–	
			Laibing	25	97	93	1.34	–	–		25	<LOD	14	32	–	–	
			Tangtang	29	110	106	1.35	–	–		29	<LOD	12	31	–	–	
			Yangchang	14	117	112	1.37	–	–		14	<LOD	0	0	–	–	
	Smokeless		RSXZ	24	103	100	1.29	–	–	–	24	62	903	100	–	–	–
Fuyuan	Smoky	Overall		81	115	108	1.41	**<0.001**	–	–	81	27	221	54	**0.0019**	–	–
		Coking Coal							**0.0044**	–					–	0.35	–
			Daping	9	93	88	1.41				9	47	54	67			
			Enhong	10	141	137	1.26				10	55	103	70			
			Haidan	6	143	139	1.32				6	<LOD	214	33			
		1/3 Coking Coal						–	0.75	–					–	0.27	–
			Bagong	8	130	125	1.37				8	1794	1486	100			
			Dahe	3	123	116	1.59				3	82	97	67			
		Gas fat coal						–	–	–					–	–	–
			Housuo	37	96	93	1.30				37	<LOD	7	32			
			Qingyun	0	–	–	–				0	–	–				
		Meager Lean Coal															
			Gumu	2	86	86	1.08	–	–	–	2	26	9	50	–	–	–
	Smokeless		Laochang	63	151	136	1.59	–	–	–	63	1065	1404	94	–	–	–

AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation; %Detect, detection rate; <LOD, values below the limit of detection (LOD).

Significant values are indicated in bold.

[a] Number of measurements is from households which exclusively burn smoky coal and report a coal source consistent with reported coal type.

[b] P-values from ANOVA test, comparing between smoky coal subtypes within each county.

[c] P-values from ANOVA test, comparing between coal source within smoky coal subtype in each county.

[d] P-values from ANOVA test, comparing coking coal emissions between Xuanwei and Fuyuan.

[e] Non-detects were replaced by LOD/squareroot(2). IQR was calculated by taking the difference between the 3rd and 1st quartile.

[f] P-values from Fisher's exact test, comparing detected/non-detected SO_2 between smoky coal subtypes within each county.

[g] P-values from Fisher's exact test, comparing detected/non-detected SO_2 between coal source within smoky coal subtype in each county.

[h] P-values from Fisher's exact test, comparing detected/non-detected SO_2 of coking coal emissions between Xuanwei and Fuyuan.

Shanghai, Shenyang, and Wuhan) showed that the highest SO_2 concentration in kitchens with coal stoves could reach up to 860 μg/m^3 (Qin et al., 1991), which is lower than the mean SO_2 concentration (907 μg/m^3) among smokeless coal users in our study. In the international context, the indoor NO_2 concentrations from our study are higher than the indoor levels measured in Indian homes using a Handy Low Volume Air Sampler, where most families were using biomass fuels for cooking (mean ± standard deviation, mean NO_2: 31 ± 24 μg/m^3 vs. 124 ± 48 μg/m^3 in our study; Kumar et al., 2008).

Interestingly, smokeless coal was associated with higher levels of SO_2 than smoky coal, even though the latter is typically considered more hazardous to human health by virtue of its role in the lung cancer epidemic in the area (Mumford et al., 1987; Lan et al., 2008). Therefore, despite smokeless coal being considered a safer alternate to smoky coal, it is potentially harmful by generating higher levels of SO_2, which has been linked to increased risks of respiratory diseases (Bruce et al., 2000). This is reflected in previous findings of higher age-adjusted mortality rates of pneumonia and risk of pneumonia death among smokeless coal users than smoky coal users (Shen et al., 2009). A prior report on coal samples collected from the same study population found significantly higher median sulfur levels in smokeless coal than in smoky coal (1.0% in smokeless coal vs. 0.2% in smoky coal, $P < 0.001$) (Downward et al., 2014a). Further, indoor SO_2 is moderately to highly correlated with sulfur levels in coal in this study population (Spearman correlation coefficient = 0.53, $P = 3.81 \times 10^{-11}$), indicating that the sulfur content of coal greatly determines the amount of pollutants released into the air. NO_x emission factor from a simulated household fire pit has also been shown to be significantly correlated with nitrogen content in coal (correlation = 0.88, $P < 0.001$) (Tian et al., 2008). An additional aspect of coal analysis was the determination of moisture content, which has been reported previously to be associated with NO_2 levels. However, we did not identify any association between the moisture content of coal and our NO_2 measurements.

In addition, our mixed effect model results showed that both number of hours spent using stoves and spring/summer season were significant positive predictors of household NO_2 concentrations. Evidently, longer duration of stove use is associated with higher

Table 3 Significant determinants of indoor NO_2 and SO_2 air concentrations (log-transformed) from mixed models

	NO_2[a]				SO_2[b]		
Characteristics	Estimate	95% CI	GMR[c]	95% CI	Estimate	*P*-value	OR
Stove ventilation							
Ventilated stove	Reference	–	–	–	Reference	–	–
Unventilated stove[a]	0.058	−0.082, 0.20	1.06	0.92–1.22	**2.27**	**0.023**	**9.63**
Mixed ventilation stove	**0.14**	**0.047, 0.23**	**1.15**	**1.05–1.26**	**1.19**	**0.020**	**3.27**
Portable stove	0.060	−0.051, 0.17	1.06	0.95–1.19	1.02	0.14	2.78
Firepit	**0.19**	**0.041, 0.33**	**1.21**	**1.04–1.40**	**2.00**	**0.041**	**7.40**
Fuel type[d]							
Smokeless	–	–	–	–	Reference	–	–
Smoky	–	–	–	–	−0.44	0.63	0.65
Plant	–	–	–	–	**−5.21**	**0.0057**	**0.0055**
Wood	–	–	–	–	**−2.66**	**0.034**	**0.070**
Other coal	–	–	–	–	−0.38	0.69	0.69
Other fuel	–	–	–	–	−0.81	0.38	0.44
Season[e]							
Autumn	Reference	–	–	–	–	–	–
Spring/summer[f]	**0.13**	**0.040, 0.21**	**1.14**	**1.04–1.24**	–	–	–
Winter	0.0054	−0.098, 0.11	1.01	0.91–1.11	–	–	–
Number of hours burning fuel[e]	**0.015**	**0.0096, 0.020**	**1.01**	**1.01–1.02**	–	–	–
Variance explained (%)							
Between subjects	16.7				24.4		
Between villages	43.0				36.7		

GMR, geometric mean ratio; CI, confidence interval; OR, odds ratio.

Significant values are indicated in bold.

[a] Estimates were obtained from linear mixed model, adjusting for variables that contributed to the lowest Akaike information criterion (AIC).

[b] Mixed logistic regression models were used, with outcomes as above or below LOD, adjusting for variables that contributed to the lowest Akaike information criterion (AIC).

[c] $GMR = \text{geometric mean ratio} = GM(\text{estimate})/GM(\text{reference}) = \exp(\beta)$.

[d] Fuel type did not significantly contribute to the prediction of NO_2 and was therefore excluded from the final model.

[e] Season and number of burning hours did not significantly contribute to the prediction of SO_2 and were therefore excluded from the final model.

[f] Spring and summer were combined due to small numbers.

indoor NO_2 concentrations. Similar to our findings, the highest indoor NO_2 concentrations were previously observed in the spring season in a study conducted in Ashford (United Kingdom) and Barcelona (Spain), which might be attributed to the contribution of outdoor NO_2 (Garcia Algar et al., 2004).

This is the first report of indoor NO_2 and SO_2 in this population. Strengths of our study include repeated visits of the same household to ensure data consistency and collection of information on multiple potential determinants affecting pollutant levels. However, we have limited power to detect differences for some smoky coal subtypes such as gas fat and meager lean coal, due to the small number of individuals using these coal subtypes. It is important to note that the number of subjects using smokeless coal is small relative to smoky coal in this region, and therefore, the smokeless coal results should be interpreted with caution. Another limitation is the lack of detailed information on smokers in each household. However, virtually all men (>90%) in the two counties smoke and therefore, any impact of smoking should be fairly uniform across the study population (Barone-Adesi et al., 2012). Further, previous research has demonstrated that tobacco has a minor impact on indoor NO_2 (Wang et al., 2012; Gao et al., 2014) and it is reasonable to consider that pollutant levels from fuel combustion should overwhelm those produced from tobacco smoke; hence, it is highly unlikely that NO_2 and SO_2 levels from environmental tobacco smoke would have a substantial impact on our findings.

In conclusion, indoor NO_2 and SO_2 concentrations vary by coal type, smoky coal source, and stove design. Future risk assessment of these pollutants should take these variables into account to ensure an accurate estimate of personal exposure and association with disease outcomes. Interventions should also simultaneously target both changing to clean fuel sources such as gas and electricity and optimizing stove ventilation. Understanding the differential NO_2 and SO_2 emissions of solid fuel types is crucial to elucidate the air components responsible for the excess of malignant and nonmalignant respiratory disease rates in this region.

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Conflict of interest

The authors disclose no potential conflicts of interest.

Supporting Information

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

Table S1. Covariates considered as fixed effects in the mixed models.

Table S2. Characteristics of the study population in Xuanwei and Fuyuan, 2008–2009.

Figure S1. High and low stoves in Xuanwei and Fuyuan, China.

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