Cancer Risk from Gaseous Carbonyl Compounds in Indoor Environment Generated from Household Coal Combustion in Xuanwei, China

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Abstract

Airborne carbonyls were characterized from emitted indoor coal combustion. Samples were collected in Xuanwei (Yunnan Province), a region in China with a high rate of lung cancer. Eleven of 19 samples (58%) demonstrated formaldehyde concentrations higher than the World Health Organization exposure limit. Different positive significant correlations between glyoxal/methylglyoxal and formaldehyde/acetaldehyde concentrations were observed, suggesting possible different sources characteristics. A sample in highest inhalation risk shows 29.2 times higher risk than the lowest sample, suggesting different coal types could contribute to the variation of inhalation risk. Inhabitants in Xuanwei also tend to spend more time cooking, and more days per year indoors than the national average. The calculated cancer risk ranged from 2.2-63 x 10^{-5}, which is within acceptable risk levels. However, cumulative effect in combination with different carbonyls could have contributed to the additive actual inhalation cancer risk. There is a need to explicitly address the health effects of low and environmentally relevant doses, considering life-long exposure in indoor dwellings.

Keywords:

Carbonyl, Indoor Air, Coal, Cancer Risk

1. Introduction

Coal is a major energy source. Coal combustion accounts for ~25% energy consumption worldwide (Zhang et al. 2008) and the trend for coal utilization is continuing to increase (i.e. annual growth of 2-3%) (Energy Information Administration 2006). China is a large energy consumer and over 75% electricity supply comes from coal combustion (Liu et al. 2008). The country is facing severe carbonaceous aerosol pollution (~ 70% due to coal combustion) (Cooke et al. 1999) and suffering from deteriorated environmental quality.

Xuan Wei County is located in the Yunnan Province of China with population approximately 1.2 million living in an area of 6,257 km^2. The county is one of the major coal producing region in Yunnan and renowned for exceptionally high lung cancer rate (i.e. 2004-05 lung cancer mortality rate: Xuan Wei; 91 per 100,000, national average; 31 per 100,000) (Lin et al. 2015). Past studies linked lung cancer mortality with coal combustion emissions in the area (Barone-Adesi et al. 2012; Kim et al. 2014; Mumford et al. 1993). The relevant coal burning studies usually concentrated on characterizing polycyclic aromatic hydrocarbons (PAHs), mineralogical compositions, household fire pit for the emissions and etc. (Chuang et al. 1992; Dai et al. 2008; Mumford et al. 1995; Tian et al. 2008). However, the role of carbonyl compounds in the coal smoke has been largely overlooked.

Airborne carbonyls (aldehydes and ketones) have been the attention of atmospheric scientists over the past few decades. Carbonyl compounds are identified with natural and anthropogenic sources. The compounds can further be produced via primary and secondary source formation such as incomplete combustion of fossil fuels and biomass, industrial emission, vehicular exhaust and photochemical oxidation of atmospheric hydrocarbons (Atkinson 2000; Carlier et al. 1986;

Grosjean et al. 2002; Kean et al. 2001; Lee et al. 1997; Perry and Gee 1995; Yokelson et al. 1999). The lifetimes of airborne carbonyls are short in troposphere (De Smedt et al. 2008; Wert et al. 2003) but nevertheless are able to undergo rapid photolysis and generate significant amount of free radicals and precursors responsible for air pollution (e.g. Secondary organic aerosol (SOA) and ozone (O_3) formation) (Carter 1994). Several carbonyl compounds are widely accepted as toxic air contaminants and contain potential carcinogenic and mutagenic properties (CEPA 1993; McLaughlin 1994; NCR 1981; Pal et al. 2008; Seco et al. 2007; WHO 2000).

Formaldehyde is a human carcinogen (Group 1) (IARC, 2006) and poses nasopharyngeal cancer (IARC 2004). Repeated occupational exposure to formaldehyde in chemical factory could increase opportunities of having health implication such as congestion in cornea, nasal membrane and pharynx (Zhang 1999). Acetaldehyde is a suspected human carcinogen (Báez et al. 2003; Zhang et al. 1994).

Indoor carbonyls concentrations is of a concern as people spend over 80% lifetime at indoor environment (Klepeis et al. 2001). Cooking and heating often involve with impure solid fuels usage in underprivileged areas (e.g. Xuan Wei) in China. The combustion processes in household coal stoves usually generate gaseous pollutants (e.g. formaldehyde, CO, CO_2, NO_x and volatile organic compounds (VOCs)) that are subject to indoor air pollution (Zhang and Smith 1999). The present study deduces possible associations between exposure to indoor coal carbonyls emissions and excess lung cancer risks at Xuan Wei, where in agony of abnormal lung cancer mortality rate in many years.

The aim of this study is to: 1) characterize gaseous phase carbonyl compounds from nineteen types of coal used in Xuanwei; 2) determine and characterize relationships between carbonyl compounds; 3) estimate the potential health implications of interior coal emissions for local inhabitants.

2. Materials and Methods

2.1 Experimental Procedures

2.1.1 Indoor Environmental Conditions Mimic "Pre-1990" Kitchen Design

Nineteen types of coal were tested for emissions. The samples were labeled 1-19 and were collected from different locations as noticed in Table 1. The samples collected from different coal seams were denoted in brackets (Table 1). Sample 7 and 8 were classified without coal seams as the samples were re-processed from coal lumps collected from the surface in the communities. Sample 10 and 12 were collected from different coal mines. The coal combustion experiment was conducted between November 2012 and January 2013, in a separate kitchen area opposite a one story building in a village called Shangzuosuo (Xuanwei). All doors and windows in the living room were closed during the experiment. The volume of the kitchen was ~42.6 m^3 (5.9 m long × 3.8 m wide × 1.9 m high). The air exchange rate in the kitchen was continuously monitored by measuring the first-order decay of carbon dioxide using a Q-Trak™ indoor air quality monitor (model 8550; TSI, Inc., Shoreview, MN, USA). The air change rate was set as 6.9 h^{-1}.

2.1.2 Preparation of Fuels

A laboratory stove (internal diameter of 15 cm; shown in Figure S1 of Supplementary Material) was used to simulate a fire pit for routine daily burning of coal. Larger coal pieces were sieved to retain only samples <5 cm in diameter, to facilitate combustion performance. The stove mass (~7 kg) and coal masses sampled (0.8 ± 0.7 to 1.6 ± 0.1 kg) were monitored throughout the experiment. The coal masses were a random factor and epitomized the usual mass range used for domestic cooking activities.

2.1.3 Burning Cycle

Nineteen types of coal samples were used in the combustion tests to collect air particulate samples. Analysis of samples of each type of coal was done in triplicate. The fire was set and kindling was ensured. The air was then purged through a stove inlet to provide oxygen for combustion and a chimney was installed over the stove to optimize the chimney effect. When full kindling had occurred (~5 min after initial ignition), ~2 kg of the coal sample was immediately added to the stove. After 10 min from initial ignition, the remainder of the coal sample was used to fill up the stove. The stove was immediately positioned above the burning coal and remained in place until completion of the experiment. The weight of the stove and coal samples were recorded. All biomass materials were completely removed outdoors, prior to setting the fire. A water pot containing 2 kg of water at room temperature was placed above the stove. Coal lumps could melt and coagulate during combustion, which could extinguish the fire. To simulate cooking in the best possible manner, the fire was stoked and poked at the beginning and at 20 min intervals during the combustion cycle to assure favorable air ventilation through the coal lumps. Additional coal was added to the stove at 20 min intervals, and the weights were recorded throughout the cycle. The water was heated to a boil during the heating process. The complete heating process required 30–60 min depending on the different types of coal (Supplementary Materials: Table S1). The remaining ashes were weighed after each combustion cycle. The combustion cycle was on par with household coal burning activity in Xuanwei (~1 h). The fire was either re-used (for another burning cycle with the same type of coal) or extinguished using a water sprayer. The weight of the coal and water was recorded at 10 min intervals during the experiment.

2.2 Samples Collection

The air samples were collected in silica cartridges impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Sep-Pak DNPH-silica, 55–105 µm particle size, 125 Å pore size; Waters Corporation, Milford, MA) at a flow rate of 0.7 L min^{-1} using an ATEC 8000 cartridge sampler. Collection efficiencies were confirmed in the field by sampling carbonyls in two identical cartridges connected in series. Efficiencies were calculated as 100% ($1-A_b/A_f$), in which A_f and A_b denote the amount of carbonyls collected in the front and back sampling tubes, respectively. No breakthrough was observed in the sampling flow rate and time used. The sampling flow rates were checked in the field at the start and end of each sampling period using a calibrated flow meter (Gilibrator Calibrator; Gilian Instruments, W. Caldwell, NJ). A Teflon filter assembly (Whatman, Clifton, NJ) and ozone scrubber were connected to the front of the DNPH-silica cartridge to remove any particulate matter and prevent possible contamination by ozone (Spaulding et al. 1999). Collocated samples were collected to testify sample collection reproducibility (>95%) in the field. A cartridge was reserved for field blank analysis during each sampling campaign and was handled in the same manner as the other sampling cartridges. The amount of carbonyls detected in the cartridges was corrected for the field blank before conversion to air concentration of carbonyl units. The DNPH-coated cartridges were stored in a refrigerator (<4°C) prior to analysis.

2.3 Carbonyls Analysis

A total of 19 carbonyls were quantified, including formaldehyde (C1), acetaldehyde (C2), acetone

(acetone), propionaldehyde (nC3), methyl ethyl ketone (MEK), butyraldehyde/isobutyraldehyde (iso+nC4), benzaldehyde (benz), isovaleraldehyde (iso-C5), valeraldehyde (nC5), *o*-tolualdehyde (o-tol), *m*-tolualdehyde (m-tol), *p*-tolualdehyde (p-tol), hexaldehyde (C6), and 2,5-dimethylbenzaldehyde (2,5-DB), heptaldehyde (C7), octaldehyde (C8), nonaldehyde (C9), glyoxal (gly) and methylglyoxal (mgly). Unsaturated carbonyls such as acrolein and crotonaldehyde were detected but not reported because of their low abundances. Unsaturated carbonyl DNP-hydrazones can react with excess reagent to form adducts, leading to ambiguities in quantification due to chromatographic interferences (e.g., double peaks) and response factor issues (Ho et al. 2011; Schulte-Ladbeck et al. 2001). In-house laboratory experiments demonstrated that collection efficiencies were >93±5% for all target carbonyls under the same flow rate, relative humidity, and temperature. Collection efficiencies for heavy carbonyl compounds (e.g., C6) were recorded to be >96±3%. Each DNPH-coated cartridge was eluted with 2.0 mL acetone-free acetonitrile solution (HPLC/GCMS grade, J&K Scientific Ltd., Ontario, Canada) and transferred to a volumetric flask. Previous studies demonstrated that neither DNPH nor DNPH derivatives remained in the cartridge after elution with 2.0 mL acetone-free acetonitrile solution (Ho et al. 2007). Certified calibration standards for monocarbonyl DNP-hydrazones were purchased from Supelco (Bellefonte, PA) and diluted to a concentration range of 15–3000 $\mu g\ mL^{-1}$. The calibration solutions were allowed to rest at room temperature for six hours for complete derivatization. The final volume of each calibration solution was filled up to 2.0 mL with acetonitrile/pyridine (HPLC/GCMS grade, Sigma) at a concentration ratio of 8:2 (v/v). The calibration curve was linearized, and the correlation of determination (r^2) was >0.999. The calibration standards and cartridge extracts were analyzed by injecting 20 μL of the solution into a high-pressure liquid chromatography (HPLC) system (Series 1200; Agilent Technology, Santa Clara, CA) coupled with a photodiode array detector (DAD). A reversed-phase separation column (4.6 × 250 mm Spheri-5 ODS 5 μm C-18, PerkinElmer, Norwalk, CT) was installed in the HPLC system and operated at room temperature (25°C). The mobile phase consisted of three solvent mixtures: mixture A, 6:3:1 (v/v) of water/acetonitrile/tetrahydrofuran; mixture B, 4:6 (v/v) of water/acetonitrile; and mixture C, acetonitrile. The gradient program was operated first at (80% A)/(20% B) for one minute, second at a linear gradient of (50% A)/(50% B) for eight minutes, third at (100% B) for ten minutes, fourth (100% C) for six minutes, and finally at (100% C) for five minutes. The elution rate was 2.0 mL min^{-1}. The absorbance of the 360 and 390 nm wavelengths was applied to identify aliphatic and aromatic carbonyls (e.g., benzaldehyde and tolualdehyde), respectively. Identification and quantification of carbonyl compounds were based on retention time and peak area integration of different carbonyl compounds. The minimum detection limit (MDL) was estimated by analyzing a minimum of seven replicates of standard solution containing analyte at a concentration of 0.015 μg mL^{-1}. The following equation was used to estimate the MDL:

$$MDL = t_{(n-1,1-\infty=99\%)} \times s \tag{1}$$

where $t_{(n-1,1-\infty = 99\%)}$ is the student's t-distribution value at n-1 degrees of freedom and S is the standard deviation of the replicates. The MDLs of the target carbonyls range from 0.002 to 0.010 ng μL^{-1}, which can be translated to 0.016–0.12 ppbv at a sampling volume of 2.02 m^3. Measured values, precision, accuracy, and validity were optimized throughout the measurements. Quality assurance was performed to ensure the above attributes were within acceptable limits. A quality control procedure was included to assure a measurement precision of 0.5–3.2% for the measured carbonyls.

2.4 Exposure Assessment and Risk Characterization

Residents living in the area are potential receptors of airborne carbonyls. Cancer risk due to exposure to gaseous phase carbonyls was estimated by considering direct inhalation exposure of inhabitants in indoor environment according to the human health evaluation manual supplemental guidance for inhalation risk assessment (Part F) (U.S.EPA 2011). The cancer risk (CR) of carbonyl compounds can be calculated by the following equations:

$$\mathrm{CR} = \text{slope factor x LADD} \quad (2)$$

$$\mathrm{LADD} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{AF} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT} \times \mathrm{CV}} \quad (3)$$

where, LADD (mg $\mathrm{kg}^{-1}\mathrm{day}^{-1}$) is the lifetime average daily dose, C (mg m^{-3}) is the pollutant concentration, IR is the average inhalation rate ($\mathrm{m}^3\ \mathrm{hr}^{-1}$). AF (%) is the absorption fraction (assume 100% absorption) (Cheng et al. 2015). EF is the exposure factor and determined by average duration in indoor (hours day^{-1}), average indoor exposure frequency (days) and average life expectancy (years). BW is the average body weight (kg). AT (days) is the average exposure duration for carcinogenic/non-carcinogenic effects. An estimated average exposure duration of 25,550 days (70 years) for carcinogenic effect is applied for the calculation, respectively (Hoddinott and Lee 2000). CV is a conversion factor (from μg to mg). The IR, EF and BW were calculated based on the information given in the Chinese exposure factors handbook and the average duration in indoor was assumed based on time-activity patterns of cooking status at kitchens in a relevant study at China (Duan 2015; Jiang and Bell 2008) . In China, population in various locations (e.g., inland versus coastal) have different economic conditions, dietary habits and living styles, thus location and region is an exposure condition that cannot be ignored. Further information can be referred to Table 2. The slope factor in equation (2) is determined by reference dose (RFD, ((mg $\mathrm{kg}^{-1}\mathrm{day}^{-1})^{-1}$)) for all carbonyl compounds according to the Integrated Risk Information System (U.S.EPA 2015). Only formaldehyde (slope factor = 0.021 (mg $\mathrm{kg}^{-1}\mathrm{day}^{-1})^{-1}$)

and acetaldehyde (slope factor = 0.01 $(\mathrm{mg\,kg^{-1}day^{-1}})^{-1}$) are considered as carcinogenic substances and therefore provided with slope factors in all measured carbonyl compounds. The CR value in a range of $1–100\times10^{-6}$ is deemed in either acceptable (10^{-6}) or tolerable (10^{-4}) level for regulatory purposes (Hu et al. 2012).

2.5 Statistical Analysis

All the data were analyzed using SPSS statistic 21.0 (IBM ®, New York, NY) or GraphPad Prism software (Version 5 for Windows).

3. Results and Discussion

3.1.1 Concentrations of Carbonyl Compounds

Table 1 shows the total and individual concentrations of carbonyls compounds in different coal samples. The formaldehyde concentrations are in a range of 10.4±5.9–502.6±148.8 μg m^{-3}. Concentrations for the acetaldehyde range from 17.0±5.9 to 195.4±40.0 μg m^{-3}. According to the World Health Organization (WHO) guideline for indoor formaldehyde is a 30-min average of 100 μg m^{-3} (WHO 2010). A total of 19 samples were analyzed: 11 (58%) demonstrated formaldehyde concentrations higher than the exposure limit. A previous study showed formaldehyde concentrations ranged from 240 to 600 μg m^{-3} in an indoor cigarettes combustion experiment (Grimaldi et al. 1996). Typical indoor formaldehyde and acetaldehyde concentrations could be in a range of 10-50 and 5-20 μg m^{-3}, respectively (Sarigiannis et al. 2011). The concentration levels in present study are akin to the combustion experiment. Formaldehyde is the most abundant compound in sample 1-7, 9-12 and 16 accounting for 21–45% of the total measured carbonyls. Acetaldehyde is nevertheless the most abundant compound in sample 8, 13-15 and 17-19 accounting for 16–33% of the total measured carbonyls. The results are consistent with formaldehyde and acetaldehyde as the dominant components in a barbecue charcoal combustion study, and also indicates concentration patterns of these carbonyls could be associated with inhomogeneous nature of the combustion raw materials (observed high standard deviation of concentrations in some of the sub-samples) (Kabir et al. 2010). A residential coal combustion study in China also demonstrated formaldehyde and acetaldehyde were the most abundant carbonyls in 5 types of coal (Feng et al. 2010). A study compared carbonyls emissions using different fuels in diesel engine showed aldehyde emissions were formed by incomplete oxidation of hydrocarbons. The formaldehyde was most abundant compound and accounted for over 40%, and the next abundant acetaldehyde ranged from 10 to 30% in composition, which have similar composition characteristics with present study (He et al. 2009). According to the U.S. Environmental Protection Agency (EPA), both formaldehyde and acetaldehyde are classified as Group B1 and B2 probable human carcinogens, respectively. The results in this study show a large proportion of potentially carcinogenic carbonyls in emissions, indicating that control is required.

Figure 1 shows correlations between the log-transformed concentrations of formaldehyde and acetaldehyde, glyoxal and methylglyoxal. Both shows positive significant correlations ($p<0.05$), however, only glyoxal and methylglyoxal demonstrates correlation coefficient > 0.80 ($n = 60$). The linear relationship between log-transformed concentrations of individual glyoxal and methylglyoxal suggests quantitative dependence of the glyoxal on methylglyoxal, which also implies the two compounds are from the same source regardless of sample locations and types. The regression analysis shows variation of glyoxal and methylglyoxal concentrations depend on the same combustion conditions (e.g. moisture content, amount of oxygen supply, temperature and etc.). A lower correlation coefficient >0.40 (n = 60) is found between log-transformed concentrations of individual formaldehyde and acetaldehyde. Carbonyl compounds could exist as reaction products from reactions of primary emitted pollutants with ozone. Secondary production of formaldehyde could be sufficient to affect indoor air concentrations. (Knudsen et al. 2003; Nazaroff and Weschler 2004; Uhde and Salthammer 2007). A past study showed relative humidity could affect the formaldehyde emissions (Parthasarathy et al. 2011). These could be possibly altering the formaldehyde concentrations and hence the concentration ratios. Nonetheless, the limitation in the correlations is that how individual combustion parameters (e.g. temperature) correlate to the concentrations cannot be determined due to insufficient combustion condition information.

The concentration ratios (C1/C2) further shows 60% samples are formaldehyde emissions dominant over acetaldehyde emissions (>1). The average concentration ratios are in a range of 0.1-3.7. The C1/C2 ratio is a common tool for characterizing pollution sources (Hedberg et al. 2002), the present trend may reflect a variety of contributing factors (e.g. temperature, relative humidity, different coal types, combustion conditions and sampling procedures), rather than taking account into individual factors alone, could all play different roles altogether in the overall variable outcome.

Strong contribution of original biogenic compounds in the lignite within early stage of coal formation could ultimately increase the coal rank (Meyer et al. 2014; Püttmann and Schaefer 1990). A previous study suggested coal combustion process could be divided into three stages: initial stage (moisture evaporation and chemical absorption), combustion stage and burnout stage-which were classified based on weight and heat changes. Thermogravimetric and differential thermal analysis showed low-rank coals could influence ignition temperatures, whereas high-rank coals influenced the burnout temperature (Moon et al. 2013).

Future studies should be concentrating on characterizing the coal materials (maturity) and different stages of carbonyls emissions such as using Proton-Transfer-Reaction Mass-Spectrometry (PTR-MS). This technique enables real-time monitoring and is able to instantaneously detect and quantify the emissions, leading to a more thorough understanding about the combustion processes.

3.1.2 Carbonyls Emissions from Various Emission Sources

Many studies targeted characterizing carbonyls emissions under different emission circumstances. A residential fireplace wood combustion study showed aliphatic aldehydes were major contributors to the gas-phase emissions from wood combustion. Acetaldehyde was emitted at highest rate among all carbonyls and the formaldehyde was at the second highest (Schauer et al. 2001). Another residential wood (softwood and hardwood) combustion emissions study suggested formaldehyde and acetaldehyde were the most abundant lower molecular weight carbonyl compounds arising primarily from the combustion of cellulose (McDonald et al. 2000).

A previous study collected samples in two residential kitchens during cooking period (used towngas and liquefied petroleum gas). The formaldehyde concentrations were 60.4 and 151 .0 µg m^{-3}. Concentrations for the acetaldehyde were 65.9 and 4.5 µg m^{-3}, respectively (Huang et al. 2011). A study measured formaldehyde and acetaldehyde levels in Paris dwellings from potentially different sources in 61 flats with no previous history of complaint for olfactory nuisance or specific symptoms. The result showed average formaldehyde and acetaldehyde concentrations ($n = 61$) in the kitchen were 21.7±1.9 and 10.1±1.8 µg m^{-3} (Clarisse et al. 2003). A past study targeted domestic levels of formaldehyde in kitchens in 185 homes at Perth, Australia with mean concentration of 25.9 μg m^{-3}. The result did not exceed the recommended Australian guideline due to good inter-room mixing of formaldehyde within homes (Dingle and Franklin 2002). A similar study measured formaldehyde concentrations in 399 home's kitchen at Ankara in Turkey showed average formaldehyde concentration was 74.9±3.7 μg m^{-3} (Vaizoğlu et al. 2003).

A study targeted different indoor areas at France (e.g. Railway station, airport, shopping center, libraries and underground parking garage). The formaldehyde and acetaldehyde concentrations ranged from 7.0-63.9 and 1.6-28.6 μg m^{-3}, respectively. Mean indoor concentrations (living room and bedroom) of formaldehyde and acetaldehyde in 16 homes were in in a range of 18.1±17.5-46.1±27.3 μg m^{-3} (Marchand et al. 2006). A study measured residences (71 homes) indoor concentrations of formaldehyde and acetaldehyde in Saskatchewan, Canada. The result suggested in both summer and winter the formaldehyde and acetaldehyde concentrations was in a range of 10.7±6.4-36.9±18.6 μg m^{-3} (Héroux et al. 2010) All of the above findings suggest the usual indoor concentrations of formaldehyde and acetaldehyde were below 100 μg m^{-3}, whereas a sample in present study showed formaldehyde concentrations from the coal emissions could be up to ~5 times, and several samples are at least ~2-3 times higher than the 100 μg m^{-3} level. The present study suggests residential coal combustion at Xuanwei could emit higher formaldehyde concentrations than ordinary indoor levels as mentioned.

3.2 Health Risk of Carbonyl Compounds via Inhalation Exposure

3.2.1 Lifetime Excess Inhalation Cancer Risk

Inhalation exposure is typically the primary route of direct exposure to airborne carbonyls. Figure 2 shows the estimated lifetime excess inhalation cancer risk (CR) per million people due to carbonyls exposure in the kitchen at Xuanwei, Yunnan Province. The non-dietary exposure in this study is defined as human exposure to gaseous carbonyls via household air. Total cancer risk value $>10^{-4}$ is considered to be at high risk in common regulatory programs (Chen and Liao 2006). Under the same carbonyls exposure condition (as in Table 1), the mean estimated excess inhalation cancer risk associated with the exposure is in a range of 22-629 cancer cases per million people (~ 2.2-63×10^{-5}) in the kitchen area at Yunnan. Formaldehyde dominated over acetaldehyde and contributed an average of ~67% of the total risk in all samples. Sample in highest inhalation risk shows ~29.2 times higher risk than the lowest sample, suggesting different coal types could contribute to the variation of inhalation risk. Under the same set of PAC emissions, the inhabitants of Yunnan show ~3.61 times higher risk compared to the national average due to different exposure conditions (Table 2) (Duan 2015; Jiang and Bell 2008) . All of the above results show the inhalation cancer risk is within acceptable (10^{-6}) or tolerable (10^{-4}) level. The carbonyls levels in the kitchen could be an important reference to other living areas in the house especially during winter as all the house windows are usually fastened with limited ventilation, in addition, inhabitants at Xuanwei spend an average of >75% of their time per day at indoors (Duan 2015). Although individual carbonyls do not demonstrate any risk under the current exposure levels, cumulative effect in combination with different carbonyls might have contributed to the actual inhalation cancer risk outcome in additive manner. The above findings suggest there is a need to revise the current risk assessment in order to explicitly address the health effects of low and environmentally relevant doses (e.g. absent of carcinogenic risk information except for formaldehyde and acetaldehyde, cancer potency factors in more than binary mixtures), considering the case of life-long exposure in indoor dwellings.

3.2.2 Limitation and Uncertainty Discussion

Many of the studies on household indoor air pollution have concentrated only on indoor air concentrations without considering personal exposure factors (Clark et al. 2013). The present cancer risk calculation is an attempt to use relevant and accessible information, as the exposure factor is specifically catered for Yunnan province and only recently launched (Duan 2015). However, the present CR calculation is not without uncertainties. A closer approximation of the actual risks could be produced if a range of weights, inhalation rates, ages and sex specific for Xuanwei inhabitants were available for the calculations. Moreover, insufficient characterization of the sampling households, for example, the number of windows and number of stoves in each household, as well as seasonal variation, could have affected the final cancer risk outcome. Furthermore, the limitation of slope factors and reference doses of several targeted carbonyls could have caused a significant under representation of the actual total risk for the analysis. Additional studies should focus on quantifying and harmonizing these uncertainties (e.g., using personal air monitoring devices to collect personal exposure data in Xuanwei households) to improve future cancer risk analyses.

4. Conclusions

The characteristics of airborne carbonyls emitted during indoor coal combustion in Xuanwei were investigated. This was 58% samples contained higher formaldehyde concentrations higher than the World Health Organization exposure limit. Positive correlations were identified in a statistical regression analysis, showing possible different sources characteristics. The lifetime excess cancer risk from inhalation of gaseous carbonyls suggests that the risk in each sample was within tolerable level. Acceptability of the risk depends on scientific data, social, economic and political factors on the perceived benefits arising from exposure to an agent.

These findings support claims that household coal combustion is associated with human health conditions. The results suggest there is a need to revise the current risk assessment in order to explicitly address the health effects of low and environmentally relevant doses.

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Table 1 Descriptive Analysis and Relative Abundances of Carbonyls Concentrations*

Sampling Location	C1	C2	acetone	nC3	MEK	iso+nC4	benz	iso-C5	nC5	o-tol	m-tol	p-tol	C6	2,5-DB	C7	C8	C9	gly	mgly	Total Carbonyls
	μg/m³																			
1 (Huchang)	305.1±311.3	162.0±115.9	71.1±43.2	27.5±19.5	43.1±26.8	15.0±7.1	88.6±62.5	20.2±11.2	20.2±12.7	16.9±11.4	46.5±32.5	19.4±13.6	23.5±19.1	15.4±14.1	15.0±9.2	11.6±7.5	15.0±8.4	18.3±17.1	12.3±11.3	946.7±636.3
2 (Wenxing)	286.3±210.6	93.7±53.9	41.6±8.3	13.0±5.2	20.8±8.0	6.7±2.4	64.0±48.9	9.3±0.8	9.7±2.7	10.6±5.1	39.4±27.7	12.9±7.1	12.4±6.3	7.9±1.3	10.5±6.1	10.0±4.3	16.3±3.1	18.6±6.3	13.2±5.4	697.1±410.0
3 (Dongshan)	201.9±161.5	130.4±59.5	60.4±23.5	25.4±5.6	43.1±13.0	14.7±3.8	70.9±40.9	27.8±3.8	32.8±8.5	18.1±4.4	40.2±13.1	18.1±5.5	19.4±5.0	12.8±3.3	17.2±8.0	16.0±3.7	20.6±7.1	7.9±4.5	8.0±5.2	785.7±360.2
4 (Tianba)	124.8±67.1	114.6±12.6	81.8±4.0	21.8±2.1	40.3±1.9	12.2±0.7	48.7±4.1	18.2±1.8	17.0±2.0	10.5±3.2	26.7±4.8	10.6±2.2	13.8±3.5	8.4±1.7	12.6±4.9	16.3±4.1	19.1±6.7	5.8±1.4	4.7±1.7	607.9±98.5
5 (Dongshan)	69.9±54.8	50.8±41.8	32.8±17.2	9.0±6.4	18.9±12.5	6.3±3.0	27.7±19.9	8.0±3.2	6.1±1.9	2.9±3.0	10.8±8.3	4.0±3.5	7.3±3.4	5.1±1.7	B.D.	7.8±0.7	10.3±1.8	3.0±1.3	2.3±1.2	283.3±184.1
6 (Jiubao)	423.0±208.5	167.1±62.5	54.7±20.5	25.5±12.2	34.8±16.1	14.8±5.8	72.4±21.2	14.5±6.4	14.7±6.7	12.4±6.9	37.8±22.4	10.3±4.2	18.1±5.8	6.8±3.8	B.D.	14.1±2.9	15.3±2.8	9.3±5.3	8.6±4.4	954.1±405.9
7 (Yefei)	502.6±148.8	195.4±40.0	72.5±26.6	30.9±5.5	39.9±9.7	18.4±5.3	87.1±26.7	20.2±9.2	22.2±12.3	18.5±3.1	46.2±9.1	17.9±2.9	20.4±6.8	11.6±6.6	10.6±3.7	12.7±3.8	13.5±3.1	10.4±6.3	12.5±6.4	1,163.6±296.4
8 (Reshui)	36.4±18.8	58.2±9.4	23.9±7.5	3.2±0.4	4.7±1.3	2.0±0.5	13.7±5.7	1.8±0.4	[a]B.D.	0.9±0.2	5.2±0.8	2.3±0.3	3.3±0.6	2.2±0.4	B.D.	9.4±2.0	5.5±0.8	0.8±0.4	1.2±0.4	174.8±16.5
9 (Laibin)	269.3±128.6	73.1±35.0	38.9±17.3	11.9±4.7	21.3±11.6	7.3±2.9	39.6±15.0	10.7±3.4	9.3±2.7	7.7±1.2	23.5±8.9	9.2±1.5	13.0±5.6	11.5±12.1	8.1±2.9	9.7±0.9	16.2±1.7	8.3±2.9	8.1±3.7	596.6±259.4
10 (Laibin)	250.1±399.8	154.7±134.9	82.3±37.1	24.8±19.8	47.9±32.7	12.1±8.2	79.4±87.4	21.1±9.0	15.5±8.8	14.3±12.9	40.1±40.5	16.4±15.7	21.1±18.8	9.1±5.2	13.9±6.7	11.0±4.8	18.6±5.9	10.2±11.0	9.9±11.0	852.7±867.8
11 (Laibin)	155.4±117.3	80.2±47.6	59.5±29.8	12.5±7.1	32.5±17.3	8.3±4.5	52.2±39.4	13.2±3.6	10.1±4.8	7.1±3.6	23.6±14.2	10.2±6.3	13.7±6.2	6.7±2.1	14.6±4.5	12.0±4.2	15.9±2.2	5.2±1.0	4.7±2.5	537.5±312.1
12 (Laibin)	122.3±20.8	96.5±31.8	74.2±25.1	17.7±4.6	38.2±8.9	8.9±1.9	41.6±6.8	19.0±5.6	13.7±2.3	7.8±1.4	22.2±4.5	9.2±2.2	13.6±3.9	7.3±2.1	16.0±9.6	11.8±4.3	19.3±3.4	8.2±2.3	6.8±2.5	554.3±130.0
13 (Zhaojiachong)	39.6±55.2	83.8±26.4	92.6±39.8	17.0±6.9	46.4±21.2	12.1±6.5	42.3±17.2	27.2±16.2	17.7±11.0	9.7±2.9	23.9±10.4	10.1±3.5	11.5±7.1	6.9±4.2	13.3±8.8	9.8±5.7	16.2±8.5	7.1±2.7	5.8±3.2	493.0±201.5
14 (Laibin)	12.3±4.8	17.0±5.9	17.6±4.1	2.4±0.7	4.3±0.3	1.6±0.3	5.6±0.3	2.3±0.6	B.D.	B.D.	2.1±0.3	1.1±0.2	2.7±0.1	1.6±0.5	B.D.	7.3±3.2	8.9±1.0	1.2±0.0	1.2±0.6	89.2±17.5
15 (Laibin)	51.0±39.8	122.4±11.5	95.0±15.3	24.4±2.6	54.5±3.0	14.3±2.5	61.9±2.9	31.1±6.0	27.5±7.5	12.5±6.0	33.1±7.7	14.6±5.5	17.0±0.8	10.8±2.7	15.0±6.6	12.8±1.5	13.3±3.0	5.0±1.5	5.0±3.1	566.3±28.8
16 (Laibin)	158.3±87.9	82.1±73.8	67.5±18.3	12.6±11.6	23.9±18.6	7.4±5.8	48.7±40.8	10.6±9.1	9.9±10.9	6.6±7.5	21.0±18.9	8.9±9.3	12.9±9.4	7.1±5.7	11.6±10.3	11.0±3.1	16.0±0.4	5.8±1.8	6.2±3.2	528.1±327.2
17 (Longchang)	57.1±48.9	90.1±34.3	75.8±24.8	12.5±4.5	27.3±10.8	7.5±2.6	38.3±15.4	9.8±2.1	7.1±1.0	3.2±1.8	15.3±6.2	5.5±2.5	8.4±1.5	4.4±2.2	7.7±1.6	10.9±0.6	13.4±0.4	4.3±0.8	3.7±1.9	402.2±152.4
18 (Laibin)	10.4±5.9	79.5±38.8	111.3±38.9	16.5±8.2	39.8±12.1	9.5±4.0	39.5±12.9	24.6±6.5	17.6±4.2	12.6±2.0	26.2±7.2	13.9±3.1	14.1±2.8	13.4±2.7	16.0±7.3	15.3±4.8	22.4±12.1	5.3±2.4	3.9±2.0	491.9±125.1
19 (Zhaojiachong)	19.2±12.2	73.6±31.4	86.2±47.4	14.5±4.2	30.9±10.1	8.5±2.5	38.8±19.3	13.7±4.6	12.8±6.7	7.9±5.9	20.2±9.5	9.5±5.4	8.7±1.6	8.5±5.4	12.1±4.0	11.0±3.1	15.4±8.4	4.9±1.9	4.0±2.0	400.4±140.7

[a]B.D. indicates below detection limit.

*n = 3 for each type of coal.

Table 2 Information about the Chinese Exposure Conditions

Exposure Factors	Yunnan	National Average by Provinces
Average inhalation rate (IR) (m^3hr^{-1})	0.645	0.654
Average duration in indoor (hrs day^{-1})	5.0	2.0
Average indoor exposure frequency (days)	320	221

Average life expectancy (years)	69.54	74.83
Average body weight (BW) (kg)	55.9	60.6
Average exposure duration (AT) (days)	25550	25550

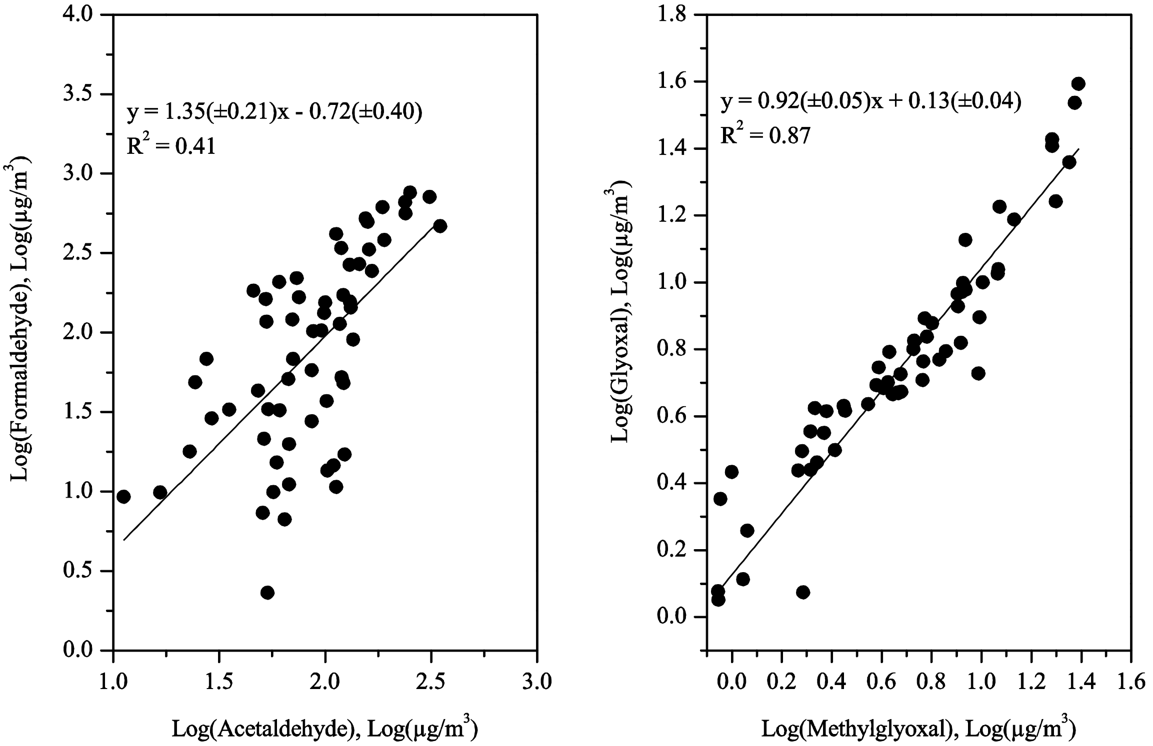

Figure 1 Relationships between log-transformed concentrations of Formaldehyde/ Acetaldehyde and Glyoxal/Methylglyoxal. There were nineteen types of coal with 3 replicates each except one type of coal with 6 replicates (n = 60). Coefficients and standard errors were included in the regression equations.

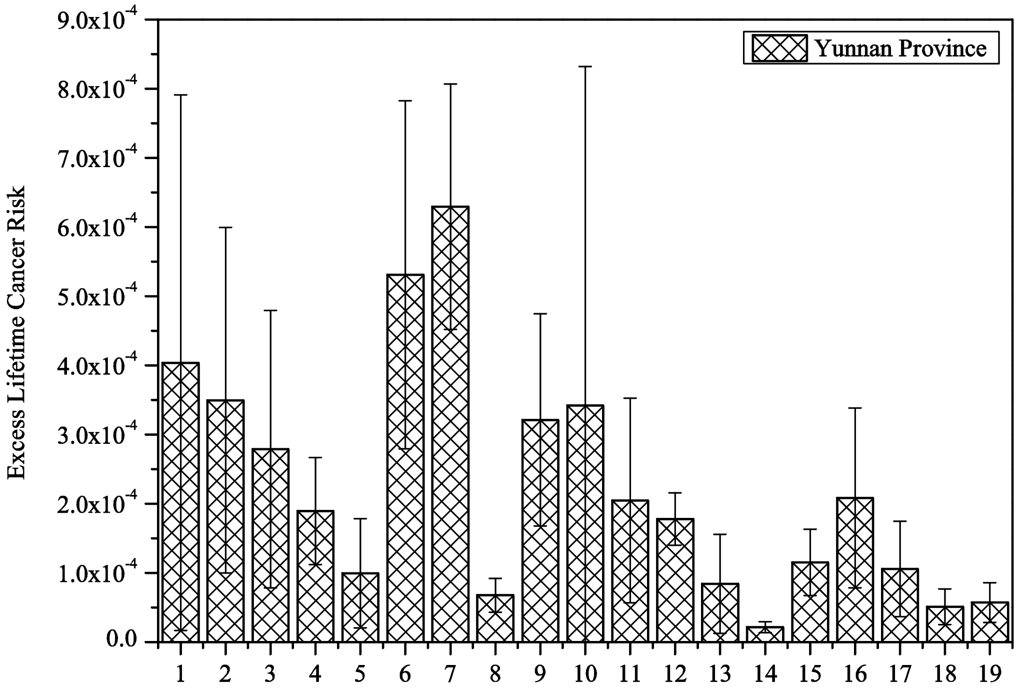

Figure 2 Excess cancer risk associated with inhalation of selected carbonyls in coal emissions. Risk error bars represent minimum and maximum values.