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Heterogeneity in coal composition and implications for lung cancer risk in Xuanwei and Fuyuan counties, China

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Abstract

Background: Xuanwei and Fuyuan counties in Yunnan Province, China have among the highest lung cancer rates in the country. This has been associated with the domestic combustion of bituminous coal (referred to as “smoky” coal). Additionally, significant geographical variation in cancer rates among smoky coal users has been observed, suggesting heterogeneity in fuel source composition and/or combustion characteristics. Research thus far has indicated that smoky coal emits high levels of polycyclic aromatic hydrocarbons (PAHs) and contains high concentrations of fine grained crystalline quartz, however, much of this research is limited in terms of sample size and geographic scope. In order to more fully characterise geochemical and elemental compositions of smoky and smokeless coal use in Xuanwei and Fuyuan, we carried out a large exposure assessment study in households in this region.

Methods: Fuel samples representing smoky and “smokeless” (anthracite, the major alternative coal type in the region) coals were collected from 137 homes in Xuanwei and Fuyuan. Rock-Eval, Leco-CS, XRF analysis and electron microscopy were used to establish hydrocarbon content

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Appendix A. Supplementary data
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(to represent volatile organic compounds), major and trace element composition and mineral composition respectively. Heterogeneity in coal characteristics between and within coal types was assessed by the Kruskal-Wallis test.

Results: 145 coal samples (116 smoky and 29 smokeless coals) were analysed. Statistically significant differences between smoky and smokeless coals with regard to hydrocarbon content, sulfur, trace elements and mineral composition were observed. Of note, smoky coal contained between 5 and 15 times the amount of volatile organic matter and twice the amount of quartz (including respirable quartz) than smokeless coal. Smoky coal generally had lower levels of trace elements (plus aluminium) than smokeless coal. Significant variation was also observed between smoky coal samples from different geographical areas with regard to hydrocarbon content and elemental composition (including aluminium and silicon).

Discussion: This paper has identified compositional differences between and within smoky and smokeless coals sourced from Xuanwei and Fuyuan counties. A decreased ratio of aluminium to silicon in smoky coal suggests elevated free silica, a finding consistent with observed higher levels of quartz. Elevated volatile organic matter content in smoky coal (when compared to smokeless coal) is consistent with the geochemical expectations for smoky and smokeless coals. These findings also reflect previous observations of elevated volatile compound emissions (notably PAHs) from smoky coal in the area. The observed heterogeneity in coal composition between and within coal types may provide leads to the observed heterogeneity in cancer risk observed in this area.

Keywords

Solid fuels; Coal; Exposure assessment; Lung cancer

1. Introduction

Xuanwei and its neighbouring county of Fuyuan, located in Yunnan Province, China, have some of the highest lung cancer rates in the nation among both males and females, irrespective of smoking status (Barone-Adesi et al., 2012; He et al., 1991; Mumford et al., 1987). Previous research has linked this excess lung cancer rate with the domestic combustion of “smoky coal” (Chapman et al., 1988; Lan et al., 2008). The term smoky coal refers to the locally available Late Permian bituminous coal (Large et al., 2009; Tian, 2005), referred to as “smoky” because of the large amounts of visible smoke released upon combustion. Coal is the primary fuel source for cooking and heating for residents in Xuanwei and Fuyuan and is available from many coal mines throughout the region. The majority of coal mines in the region produce bituminous coal while a small number produce anthracite coal (originating in the Carboniferous period (Tian, 2005) and referred to as “smokeless coal”), which is associated with a relatively lower cancer risk. For clarity, and consistency with previously published research related to lung cancer in Xuanwei and Fuyuan, we shall refer to these coals as smoky and smokeless, using the term “type” to differentiate between them.

The lung cancer rate among those who routinely use smoky coal has shown considerable heterogeneity between geographic locations (Lan et al., 2008; Lin et al., 2012). This likely reflects different styles of coal preparation (such as making coal briquettes, or packing coal dust with local clay/soil) and/or compositional differences in coal between geographic sources. These compositional differences, which may in turn reflect geological differences in coalification or depositional environment, are indicated also by the sub-categorization of smoky coal by the State Standard of China Coal Classification (Chen, 2000) into: coking coal, 1/3 coking coal, gas fat coal and meagre lean coal. The factors which drive this sub-categorization are the combination of the degree of coalification (which is measured as the dry ash free volatile matter — V_{daf}) and the caking property of the coal (represented as a combination of the caking index, the maximum thickness of the plastic layer and the Audibert- Arnu dilation — see Table 1 (Chen, 2000)).

Research on coal samples collected from mines has indicated that smoky coal contains high amounts of quartz (classified by the International Agency for Research on Cancer (IARC)) as carcinogenic to humans (Straif et al., 2009) when compared to locally sourced anthracite coal and bituminous coal sourced from other areas in China and from the USA (Dai et al., 2008b; Large et al., 2009; Tian, 2005). However, much of this previous research has been restricted to small sample sizes and has focussed upon the region in Xuanwei with the highest lung cancer rate (i.e. Laibin commune) with very limited comparison to alternative fuel types (e.g. smokeless coal), making it unsuitable for the examination of heterogeneity in lung cancer rates and coal composition. A further limitation of analysing fuel collected directly from coal mines is that it may not accurately represent the fuel to which residents are exposed if modifications are made by residents prior to combustion, for example by making mixtures of coal and clay in order to conserve fuel by extending its burning life. Research focusing upon the combustion products of smoky coal has revealed that smoky coal (when compared to smokeless coal) emits relatively high amounts of particulate matter, nanomaterials (Hosgood et al., 2012) and organic compounds, specifically polycyclic aromatic hydrocarbons (PAHs), most notably Benzo[a] Pyrene (BaP) (Mumford et al., 1987), classified by the International Agency for Research on Cancer (IARC) as carcinogenic to humans (Baan et al., 2009).

This paper presents the results of the largest analysis to date of coal samples collected directly from homes of residents of Xuanwei and Fuyuan counties. The objective of this paper is to catalogue the geo-chemical and elemental compositions of the smoky and smokeless coals that are currently used by the populations of Xuanwei and Fuyuan. Emphasis is placed upon differences between smoky and smokeless coals as well as geographic variation in the composition of smoky coal.

2. Methods

2.1. Study design

This paper forms part of a large lung cancer case-control and cross-sectional molecular epidemiology study aimed at comprehensively cataloguing the constituents and associated burning products of solid fuels used in the Xuanwei and Fuyuan counties, and ultimately linking those constituents to specific lung cancer risks. 30 villages were selected from Xuanwei and Fuyuan (15 from each). Villages were selected to provide a representative overview of the local population and a variety of coal sources. Approximately five households were selected from each village. Selection was intended to represent the case-control study population in regard to both age and gender (all participants in the case-control study are female) as well as the typical variety of living arrangements, stove types and ventilation characteristics. The selection criteria for households were:

- At least one female between the ages of 20 and 60 residing there at the time of the study
- Stove type and configuration not changed for at least the past five years
- The house is at least ten years old.

One eligible female in each household was enrolled for personal monitoring of airborne pollutants and provided biographical information (including medical histories), biological samples (blood, sputum, urine, etc.) and logged their activities during the monitoring periods. Data was collected over 2 time periods — the first being from August 2008 to February 2009 and the second being from March to June 2009. In the first time period, 149 participants, from all 30 villages were recruited. In the second, 16 of the initial 30 villages were re-visited. Villages were selected for revisiting such that a representative set of the overall fuel usage in Xuanwei and Fuyuan was represented. 53 subjects were re-sampled and 15 new subjects were recruited. During each visit, indoor and outdoor air pollution was measured and questionnaires covering personal activities and fuel usage during each measurement period were collected. Participants who reported the use of coal were also asked whether it was smoky or smokeless coal, and the mine from which it came (residents will typically purchase their coal in bulk once or twice a year). Smoky coal subtypes were established by matching the reported mines to their official coal classification as defined by the State Standard of China Coal Classification (coking coal, 1/3 coking coal, gas fat coal or meagre lean coal (Chen, 2000)).

2.2. Fuel sample collection

Fuel samples were collected from the homes of study participants. The type of fuel used (e.g. coal, wood) was recorded and, if applicable, the type (smoky/smokeless), and source of coal were recorded. 275 fuel samples were collected in total, 195 coal based samples, 64 wood and 16 “other” plant products (corn cobs, bamboo and/or tobacco stems). The coal based samples comprised solid coal, fragmented coal and manufactured coal briquettes. Only solid and fragmented coal samples were analysed for this paper. Participants did not differentiate between solid and fragmented samples and would refer to them both as “coal”. Manufactured briquettes (n = 18) were not analysed as we could not determine the origin of the coal. The total number of coal samples (solid and fragmented) analysed was 177.

2.3. Coal sample selection for analysis

Of the 177 coal samples, 137 were from unique homes. 40 homes provided 2 coal samples, 31 of which were duplicates with regard to coal type and visitation period to those already collected from their respective homes and were not analysed. Of the remaining 9 homes, 2 reported burning 2 different types of coal and 7 provided coal samples during both the first and second data collection periods (4 used the same coal type during each period while the remaining 3 had changed coal types). In these cases all coal samples were analysed. The total number of samples analysed was 146.

Samples were analysed to assess the bulk organic geochemical composition, carbon and sulfur contents and major and trace element composition.

2.4. Bulk organic geochemical analysis

The bulk organic geochemical analysis (Rock-Eval), indicating hydrocarbon generation in an inert atmosphere at 300 °C (S1), hydrocarbon generation during a programmed pyrolysis from 300 to 650 °C at a heating rate of 25 °C per minute (S2) and the temperature at which maximal hydrocarbon generation (Tmax) occurs, was carried out via standard Rock-Eval VI process (Behar et al., 2011). Coal samples were finely ground under constant pressure using a Herzog HM grinding machine but were otherwise analysed as received. S1 and S2 were used to broadly approximate volatile organic compounds and cracked hydro-carbons, respectively and were measured in milligrams of hydrocarbon per gram of coal (mg HC/g coal). Tmax was measured in degree Celsius.

2.5. Carbon-sulfur analysis

Total carbon and sulfur are measured with a LECO SC 632 using finely ground coal samples in an as received state. The total carbon content was analysed by means of total combustion of 0.1 g of each sample at 1350 °C and subsequently measuring the CO_2 and SO_2 release by means of an infra-red detector. The amount of CO_2 and SO_2 is calibrated with a pure calcite and Ag_2SO_4 standard, respectively. For accuracy, in- house geological standards are measured every 10 samples. The total inorganic carbon content is determined by a second analysis on a sample from which the organic carbon is removed by heating the material at 550 °C. The total organic carbon content is calculated as the difference between the total carbon content and the total inorganic carbon content.

2.6. Ash yield

Ash was obtained from a standard thermogravimetric analysis (TGA) using a temperature programme from 105 to 1000 °C. The ash yield is displayed as a percentage of each coal specimen.

2.7. Elemental composition

Elemental composition was established through X-ray fluorescence (XRF) on glass beads prepared from the dry ash obtained as detailed above. Results of XRF analysis were corrected for the loss of ignition (LOI) after adjustment for moisture. In order to fully explore constitutional differences between smoky and smokeless coals a wide range of elements was measured. These were: Silicon (Si), Aluminium (Al) Titanium (Ti), Iron (Fe), Manganese (Mn), Calcium (Ca), Magnesium (Mg), Sodium (Na) Potassium (K), Phosphorus (P), Chromium (Cr), Nickel (Ni), Strontium (Sr), Barium (Ba) and Zirconium (Zr). Elements were initially reported as a percentage of coal when bound to oxygen (i.e. SiO_2). Levels of the individual elements were calculated using their respective atomic masses and converted to parts per million (ppm).

Quartz content (as a percentage of coal) was estimated by the use of the following formula (Tian, 2005):

$$\text{Estimated Quartz} = (SiO_2)_{total} - 1.5 \times Al_2O_3 .$$

2.8. Mineralogical analysis

25 coal samples, (approximately one per village), representing 19 smoky coals (11 from Xuanwei, 8 from Fuyuan) and 6 smokeless coals (2 from Xuanwei and 4 from Fuyuan) underwent scanning electron microscopy (SEM) to assess quartz and other mineral content of the coal samples. The physical size of quartz grains was measured and grains less than 9.6 μm were considered to be part of the "respirable fraction" (of a size that, if inhaled, could theoretically reach the end alveoli (World Health Organization, 1999)). The results are presented as a percentage (%) of measured coal. Crystal structure was not included in the analysis.

2.9. Quality assurance

The reliability of the results in the full dataset was established by in-house testing of ISE921 standard reference samples. This indicated consistent functioning of the analytical devices. A subsample of the coal samples (n = 7 [5%] − 5 smoky and 2 smokeless) was also reanalysed, showing consistency of measurements for all tests.

2.10. Statistical analysis

Normal probability plots indicated that the results of the Rock-Eval evaluation, carbon-sulfur analysis and SEM were not normally or log-normally distributed. The results of the XRF analysis showed a log-normal distribution. Descriptive statistics are presented as medians. Wilcoxon-ranked tests were performed to assess differences between smoky and smokeless coals and Kruskal-Wallis tests were performed to assess for heterogeneity between and within designated smoky coal sub-types. Variation within smoky coal sub-types was assessed by specifying the coal mine that each coal sample was sourced from and assessed for variation between those coal mines. In testing for variation between coal types and sources, only the coal types which were consistent with their reported region were included (n = 140 [96%]). Where possible, the results were broadly compared to equivalent average values for coals from throughout China and the USA (see Table 2 (Dai et al., 2008b; Bragg et al., 1998)).

Within the XRF results, multi-correlate testing and exploratory factor analysis were carried out on the log-transformed values to identify underlying latent structures. Factor analysis was performed with varimax rotation. Factors with eigenvalues greater than one were retained. Individual variables with loading values of greater than 0.5 were considered to significantly contribute to that factor. Identified factors underwent further Kruskal-Wallis testing to assess variation in the latent structures between coal sources.

The primary focus of this paper is to assess the constituents of the whole coal samples as this is what is used by the inhabitants ofXuanwei and Fuyuan. However, to ensure that the ash yield of the coal samples was not contributing to any observed differences between coal samples (for example, if a coal specimen contained a relatively high ash yield then that may result in an under-reporting of the other coal constituents), statistical testing was repeated following standardisation for ash yield (i.e. measurements per unit ash).

In all statistical tests, a p value of less that 0.05 was used to indicate statistical significance. Statistical testing was carried out in R version 2.14 (R Development Core Team, 2011) utilising the Hmisc, reshape2, lattice and corrgram packages (Harrell, 2013; Sarkar, 2008; Wickham, 2007; Wright, 2011).

3. Results

3.1. Overview

146 coal samples were analysed. One sample was excluded after analysis revealed highly spurious results, indicative of analytical failure. Of the samples retained for analysis, 72 were from Xuanwei and 73 were from Fuyuan county, of which 68 Xuanwei samples and 66 Fuyuan samples could be matched to specific coal mines. In Xuanwei, all identified smoky coal mines were of the coking subtype (i.e. Azhi, Baoshan, Laibin, Tangtang and Yangchang). In Xuanwei, there is a region which historically produced smokeless coal (although now many residents burn wood as their primary solid fuel), referred to as “RSXZ” which is an amalgamation of the ReShui and XiZe areas. Fuyuan mines produce a variety of smoky coal subtypes. Coking coal is produced by the Daping, Enhong, Haidan and Zude mines. Bagong and Dahe produce 1/3 coking coal, Housuo and Qingyun produce gas fat coal and Gumu produces meagre lean coal. There is one smokeless coal mine in Fuyuan called LaoChang. The geographical positions of these mines and their corresponding coal types are shown in Fig. 1.

3.2. Coal reclassification

Coal subtypes (smoky or smokeless) were reported by the study participants. This reporting gave 26 smokeless, 114 smoky and 5 unspecified coal types. Rock-Eval analysis indicated that the “Tmax” measurement showed a clear demarcation of coals into two groups, one above and the other below a Tmax value of 550°. This demarcation predicted 107 of the 114 (94%) coals designated smoky and 22 of the 26 (85%) coals designated smokeless and indicated that all of the unspecified coals were of the smoky coal variety. Based on this clear demarcation, and the consistency with other reported Tmax values for equivalent coals from other sources (e.g. Laumann et al., 2011), coals were reclassified as smoky or smokeless on the basis of their Tmax measurement. In order to ensure that the results were not unduly influenced by this imputation, all statistical analyses were re-performed using only coal samples which matched with their original reported sub-type (n = 129). The results were consistent with those presented here (Tables S4 and S5 in the Supplement).

3.3. Ash yield

Analysis of ash yield (Table 3) revealed variation between the smoky coal subtypes sourced from Fuyuan, but there was no significant difference between smoky and smokeless coals or within designated smoky coal subtypes. The median ash yield for all coal samples was 32%, which is the same as the median ash yield for both smoky and smokeless coals. Some variation was observed in the median values within smoky coal subtypes, for example, ash yield within Fuyuan coking coals ranged from 23% to 51% and within Xuanwei coking coals from 23% to 36%, however this difference did not reach statistical significance.

3.4. Rock-Eval analysis

Rock-Eval analysis revealed several (expected) differences between smoky and smokeless coals (Table 3). As stated, smokeless coal has a significantly higher Tmax than smoky coal (median value 581 °C vs. 460 °C respectively, $p < 0.001$). The median values for S1 and S2 in smoky coal (2.15 mg HC/g coal and 71.5 mg HC/g coal respectively) are significantly higher (by up to a factor of 10) than those for smokeless coal (0.29 mg HC/g coal and 8.39 mg HC/g coal, $p < 0.001$). On investigating for geo-spatial variation, we observe that there is a significant variation among the five coal mines in Xuanwei designated “coking coal” in the values of Tmax (ranging from 443 to 463°°C), S1 (1.62–4.03 mg HC/g coal) and S2 (61.59–138.69 mg HC/g coal). In Fuyuan, a significant variation between the four smoky coal subtypes produced in the region for the values of S2 (43.96–131.87 mg HC/g coal) and Tmax (433–467°°C) was observed. Within the two coal mines producing 1/3 coking coal, a significant difference between the Tmax scores was observed (445 vs. 465°°C). Significant variation between the two smokeless coal areas was observed for the S2 measurement (6.49 vs. 9.47 mg HC/g coal). Standardising values by ash yield (Table S6 in the Supplement) did not alter the relative differences between smoky and smokeless coals.

3.5. Carbon-sulfur

Carbon-sulfur analysis revealed internal variation within coal types (Table 3). The median values for total (59.4%), organic (58.8%) and inorganic carbon (0.5%) for smoky coal are slightly higher than those for smokeless coal (57.4%, 56.9% and 0.3% respectively), although these differences are not statistically significant. The median value for sulfur in smoky coal (0.2%) is significantly lower than that in smokeless coal (1.0%, $p < 0.001$). On investigation for geo-spatial variation, statistically significant variation within the Xuanwei coking coals for total (range 55.76–70.12%), inorganic (0.19–0.92%) and organic (54.17–69.77%) carbon was observed. In Fuyuan, significant variation between smoky coal subtypes for total (50.83–64.34%), inorganic (0.2–1.4%) and organic carbon (53.3–63.7%) was observed. A significant difference between the two 1/3 coking coal regions in Fuyuan was observed for sulfur content (1.88 vs. 0.16%). In the smokeless coal samples there was a significant variation between the two smokeless coal producing mines for total (57.41 vs. 46.17%) and organic (45.81 vs. 57.33%) carbon and sulfur (1.03 vs. 2.83%). Both smoky and smokeless coals have lower levels of total carbon and sulfur than the general USA and China coal values (Table 3). Adjusting for ash yield (Table S6 in the Supplement) results in the loss of the previously observed statistically significant variation within Xuanwei coals and Fuyuan coking coals (implying that the ash free coal components have similar carbon contents).

3.6. Elemental composition

XRF revealed multiple differences between smoky and smokeless coals (Table 4). Median values of silicon for smoky (92,100 ppm) and smokeless (84,000 ppm) coal were both higher than that observed for China in general (39,700 ppm) and the USA (23,900 ppm).

While the median level of silicon among smoky coals was observed to be higher than that among smokeless coals, this difference did not reach statistical significance ($p = 0.21$).

In general, smoky coal was observed to have lower levels of trace and major elements than smokeless coal. Elements for which smoky coal had significantly lower levels were: Al (19,750 ppm vs. 43,800 ppm, $p < 0.001$), Ti (1,400 ppm vs. 5100 ppm, $p < 0.001$), Na (200 ppm vs. 600 ppm, $p < 0.001$), K (700 ppm vs. 4100 ppm, $p < 0.001$), P (100 ppm vs. 300 ppm, $p < 0.01$), Cr (20.4 ppm vs. 35.1 ppm, $p < 0.01$), Ba (36.8 ppm vs. 108.8 ppm, $p < 0.001$) and Zr (66.5 ppm vs. 156.7 ppm, $p < 0.01$).

On investigating for geospatial variation, we observed a significant variation within the coking coal subtype in Xuanwei for Si, Fe, Mn, Ca, Mg, Na, Cr, Ni, and Sr. Among the Fuyuan coals, a significant variation between the three smoky coal subtypes for Al, Ti, Fe, Mn, Ca, Mg, Na, K, P, Cr, Ni, Sr, Ba and Zr was observed. Variation was also observed within coals sourced from Fuyuan among: the coking coal regions for Na; the 1/3 coking regions for Fe, Ca, Cr and Sr; and among the gas fat regions for Ca and Sr. Significant variation between the two smokeless coal producing regions was observed for Al, Ti, Fe, Mg, K, Cr, Ni, Ba and Zr.

Adjusting for ash yield (Table S7 in the Supplement) resulted in some loss of the previously observed statistically significant variation within some of the smoky coal subtypes. Of note, the significant variation on Si levels within Xuanwei coking coals was lost. However, in general the observed differences between smoky and smokeless coals remain largely unchanged.

Estimated quartz content, based on the aluminium and silica content, revealed that smoky coal had a significantly higher proportion of estimated quartz than smokeless coal (median value 11% versus 5.8%, $p < 0.001$). A significant variation within the Xuanwei coking coals, ranging from 9.6% for coals from Yangchang to 17.5% for coals from Tangtang, was observed ($p < 0.05$). A significant difference in estimated quartz between the two smokeless coal producing mines was also observed with coal from LaoChang containing 6.5% estimated quartz and coal from RSXZ containing 0% estimated quartz, $p < 0.001$.

XRF results were analysed further through the use of correlograms and factor analysis. The correlograms are displayed in Fig. 2 and indicate that while there are many similarities between the internal correlation structures of smoky and smokeless coals there are some differences between the two coal types. In general smoky coal showed more and stronger positive correlations than smokeless coal. Negative correlations in smoky coal were restricted to Mn and Ca while for smokeless coal this was more diffused with negative correlations scattered between Fe, Mn and Ca with Si, Ti, Na, K, Sr and Ba. Correlograms created following adjusting for ash yield were largely identical to those presented here (Fig. S1 in the Supplement).

The results of the factor analysis are presented in Table 5. Three factors were identified in smoky coal which together accounted for 77% of the observed variance. The first factor consisted of Al, Ti, Na, K, P, Cr, Ni, Ba and Zr, a combination suggesting that this factor represents lithophile elements. The second factor consisted of Mn, Ca, and Sr, which suggested that this factor might reflect carbonate. The third factor consisted of Fe, Mg and Ni, possibly suggesting a volcanic ash deposition during coal formation. Factor analyses on the smokeless coal samples identified five factors accounting for 86% of the observed variance. The first factor loaded heavily on Al, Ti, Fe, P, Cr and Zr, which is similar as to smoky coal represents lithophile elements. The second factor loaded onto Na, Sr and Ba, which may partly represent a marine influence on the coal during deposition. The third factor loaded onto Si, and Ni, the fourth on Ca and the fifth on Mg and K. It is presently unclear what the third, fourth and fifth smokeless coal factors represent. Factor analysis following adjusting for ash yield resulted in the same factors being identified as those presented here (see Table S8 in the Supplement).

Scatter plot matrices of the factors are presented in Figs. 3 and 4. Among smoky coals, the differing smoky coal subtypes (coking coals from Xuanwei and Fuyuan are displayed separately) have been indicated. Among smoky coals, variation in factor loadings can be observed between coking coal and gas fat coal. Furthermore, a variation in the factor loadings between coking coal from Fuyuan and that from Xuanwei was observed. Statistical testing revealed a significant variation in factor loadings ($p < 0.05$) between the smoky coal subtypes for all the three factors. In the smokeless coal samples, variation in loadings of factors was observed between the two smokeless coal producing areas. Statistical testing indicated significant variation ($p < 0.05$) in the loading patterns of factors 1, 2 and 5 (but not 3 or 4) between the two areas.

3.7. Mineralogical analysis

Mineralogical composition was established on a subset of coal samples via the use of SEM ($n = 25$). Smoky coal was found to have significantly higher amounts of total quartz (4.6%) than smokeless coal (2.2%, $p = 0.03$, Table 6). This extended to quartz within the respirable fraction (1.9% versus 0.6%, $p < 0.01$). Among other minerals, carbonate (2.2% versus 0.2%, $p = 0.04$), and albite (0.4% versus 0.2%, $p = 0.05$) were present in significantly higher levels in smoky coal than smokeless. Full results for the mineralogical analysis are available in the Supplement (Tables S9 and S10).

4. Discussion

The lung cancer rate in Xuanwei and Fuyuan counties is among the highest in China. This excess risk has been linked to the domestic usage of locally sourced smoky coal (Barone-Adesi et al., 2012), with additional geographic variation in lung cancer rates observed among smoky coal users (Lan et al., 2008; Lin et al., 2012). The primary aetiological factor(s) present in smoky coal have not yet been identified. At present, the primary hypotheses implicate the emission of hydrocarbons, in particular PAHs (Mumford et al., 1989), as the major carcinogenic factor, with the possibility of mineralogical agents (specifically silica/quartz) being involved (Large et al., 2009; Tian, 2005). Previous research investigating coal samples from the region acquired coal directly from mines (Dai et al., 2008b; Large et al., 2009). This procedure, while giving highly detailed information regarding coal seam, type and precise location, is limited when trying to apply these findings to an exposure assessment context. This limitation arises because in order to accurately portray individual exposure patterns no alteration (e.g. briquetting, blending with clay, weathering) must have been made to the coal product between mining and domestic use. Our observation that residents had a broad definition of coal, which extended to both whole and fragmented coals (which, upon gross visual inspection of selected samples showed evidence of being mixed with local dirt/clay to make a coal cake or briquette) would lend considerable uncertainty to that assumption. By collecting fuel samples directly from homes we are able to accurately collect, analyse and characterise the fuel currently being used.

Many of the observed differences between smoky and smokeless coals are to be expected given the generally known differences between bituminous and anthracite coals. However, these findings support the assertions that: smoky and smokeless coals are fundamentally different; heterogeneity within smoky and smokeless coals exists; and add plausibility to the current consensus that the elevated lung cancer risk in the region can be attributed to smoky coal.

4.1. Rock-Eval analysis and the role of volatiles

The finding of higher hydrocarbon availability in smoky coal (as indicated by the hydrocarbon release at 300°°C (S1) and during a programmed pyrolysis from 300 to 650°°C (S2)) compared to that of smokeless coal is consistent with the expectation that bituminous coal would, consequently to its being a less mature coal than anthracite, have a higher potential hydrocarbon release (as indicated by S2). The relative differences in S1 measurements between coal samples are broadly consistent with the stipulations for volatile matter set by the Chinese standard of coal classification (V_{daf}, see Table 1), with the exception that the volatile matter in coking coal would be expected to be lower than that of 1/3 coking coal, but would instead appear to be approximately equivalent. The hydrocarbon content in smoky coal is between 5 and 15 times that for smokeless, which reflects previous research observing an approximately 25 fold difference in indoor BaP levels between smoky and smokeless coals (Mumford et al., 1989). This would lend some support to the hypothesis that the organic fraction including PAHs is at least in part responsible for the excess cancer risk in the region. It must be noted though, that the classes of hydrocarbons measured in the geochemical analysis are not known and that there are multiple types of organic compounds that could be represented by these values.

The Tmax measurements (temperature at which maximal hydrocarbon generation occurs) made it apparent that some (n = 11) coal types had been misclassified and needed reclassification. This decision is supported by similar Tmax measurements for equivalent coals (Laumann et al., 2011) and by our own internal sensitivity analyses where we established that the results from the updated coal types are consistent with that which would have been obtained had we excluded those coal samples.

4.2. Elemental composition and quartz

The XRF analyses of the coal samples reveals that smoky coal, in general, contains lower levels of elements when compared to smokeless coal. It is possible that the levels of several elements (e.g. Na, P, Sr) may have been under-reported due to volatisation during the ashing process at 1000°. However, we would not expect this to alter the relative differences between smoky and smokeless coals. The difference in elemental composition also extends to identifying latent variables through the use of factor analysis, which identified 3 factors in smoky and 5 in smokeless coals. Both coal types appeared to have lithophile elements (i.e. those elements which predominantly occur in silicate minerals and are concentrated in the crust) contributing to their first (and highest loading) factor, however the subsequent factors appear to be different between the coal types. Of the remaining four factors for smokeless coal, one potentially represents previous marine influence while the remaining three have no presently known geological meaning. The remaining two smoky coal factors appear to represent carbonate and volcanic influence respectively. These factors appear to be consistent with the observed geology of the area, as a research carried out upon coal samples collected from mines in Yunnan has indicated the presence of carbonate layers and volcanic influence in the formation of coal in the area (Dai et al., 2008a,b).

Previous research has indicated that the levels of silicon in smoky coal are higher than those observed in similar coals in the USA and other Chinese sources (Dai et al., 2008b; Large et al., 2009). Our study has confirmed this assertion, but has also shown that silicon in smokeless coal is at a similar level as smoky coal. If we assume that all silicon detected exist in a mineralogical capacity (i.e. as silica) and consider the relative difference in aluminium levels between smoky and smokeless coals, we would predict that there are marked differences in the mineralogical state of silica between smoky and smokeless coals. Mineralogically, silica either exists as “free” (i.e. as SiO_2 crystals - of which quartz is the most common) or combined with metals - of which aluminium is the most common (aluminosilicates). It has been postulated that the “expected” ratio between silica and aluminium in these aluminosilicates is 1:1.5, with the excess silica representing “free” silica and thus being an indicator of quartz content (Large et al., 2009; Tian, 2005). Using this premise we predicted quartz levels for the entire dataset and found significantly higher levels of estimated quartz among the smoky coal samples. This was reflected by the finding of higher quartz identified at the surface of smoky coal via SEM. While these results are numerically different from each other (estimated values are approximately double that measured by SEM) they each show a similar relative difference in quartz levels between smoky and smokeless coals (approximately double). Furthermore, correlate testing indicates that the estimated and measured quartz values are positively correlated with a Spearman correlation coefficient of 0.6. This would indicate that, in addition to confirming previous assertions of elevated quartz content in smoky coal (Dai et al., 2008b; Large et al., 2009; Tian, 2005), the use of the relative difference between silicon and aluminium provides a reasonable method for comparing relative differences in quartz between smoky and smokeless coals in this region.

Quartz is categorised as carcinogenic to humans by IARC (Baan et al., 2009) and so this finding has implications for a potential aetiological factor for the lung cancer epidemic in the region, either alone (Tian, 2005), or in combination with hydrocarbons/volatiles (Large et al., 2009). However it has been pointed out that the attributed degree of carcinogenicity to quartz is insufficient to fully explain the degree of cancer risk observed (Vermeulen et al., 2011). Furthermore, these assessments are restricted to the analysis of uncombusted coal samples. As yet, no published articles investigating quartz in combustion related indoor air pollution are available. Thus, we are unable to state whether or not these elevated levels of quartz become aerosolized during the combustion process or in the clean-up of ash.

4.3. Variability within coal types

Variation in constituents between designated smoky coal subtypes (coking coal, 1/3 coking, etc.) is to be expected to a degree as the geo-chemical properties defining the smoky coal subtypes are represented by some of the measurements presented in this paper. However, these findings provide support for previous observations of varying cancer rates between regions (Lin et al., 2012), which implies a relationship between coal area and carcinogenicity. A finding of variation between mines producing the same coal subtype (as seen for coking coal from Xuanwei) would imply either regional differences in coal preparation or further heterogeneity in coal generation within designated coal sub-types, which may assist in further identifying variation in carcinogenic constituents between locations. However, how this observed heterogeneity in coal constitution relates to personal exposure and specific cancer risks is not yet known and needs to be investigated further before drawing any aetiological conclusions.

While this paper is the most comprehensive analysis of coal from Xuanwei and Fuyuan to date, some limitations relating to sample size exist. Coal samples were taken throughout the whole of Xuanwei and Fuyuan counties, providing a wide variety of coal samples but a limited number of samples from each region. Additionally, while attention has been given to the variation in coal composition between mines (changes in the horizontal plane), this paper is unable to ascertain whether compositional changes in coal have occurred as a result of decades of mining (changes in the vertical plane). Given the long latency period of lung cancer (Pass et al., 2010), there may have been variations in carcinogenic exposures over these previous decades. However, there is some evidence to suggest that there has been no major compositional change within at least the last 2 decades as the research that has compared Xuanwei coal mined 20 years ago with recently excavated coal (Large et al., 2009) has indicated little compositional difference between the two specimens.

4.4. Summary and conclusion

The excess lung cancer risk in Xuanwei and Fuyuan has previously been linked to the domestic combustion of smoky coal with a strong geographic heterogeneity within homes that regularly use smoky coal. This paper has identified a clear difference in coal composition between smoky and smokeless coals among the aspects of hydrocarbon content (i.e. potential hydrocarbon emissions), elemental composition and quartz content. Variation is also observed within coal type (samples) dependent upon their geographical source. These results parallel heterogeneity in cancer risk resulting from the use of coal in this region.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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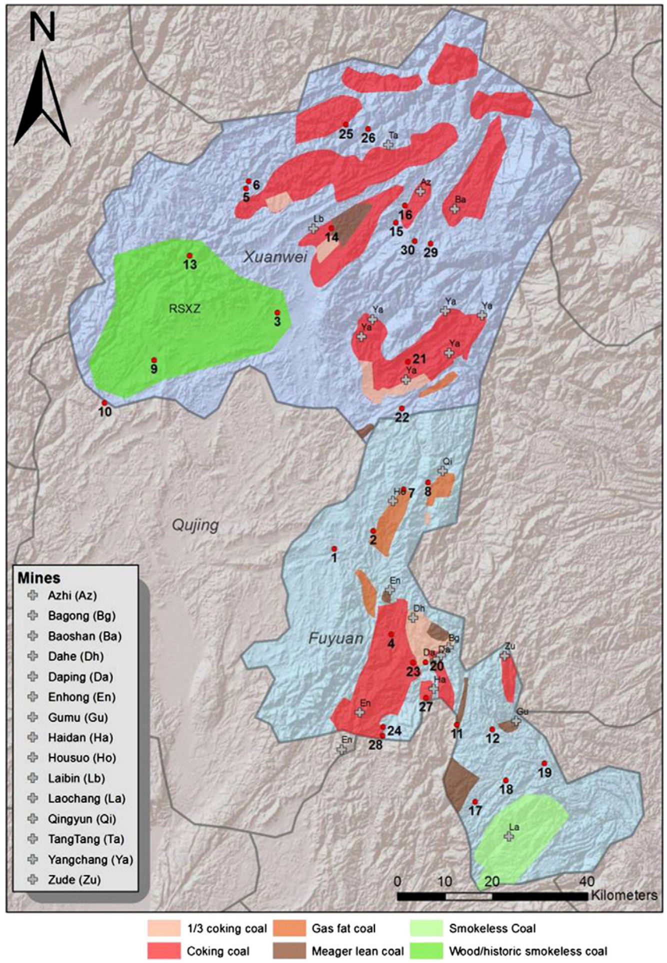

Fig. 1.

Map of Xuanwei and Fuyuan counties showing approximate locations of study villages, reported coal mines and coal sub-types. Villages enrolled in the study are marked numerically 1 to 30. The mine locations indicated represent functioning mine entrances, therefore, some mines may be indicated more than once. Only mines reported by study participants are indicated.

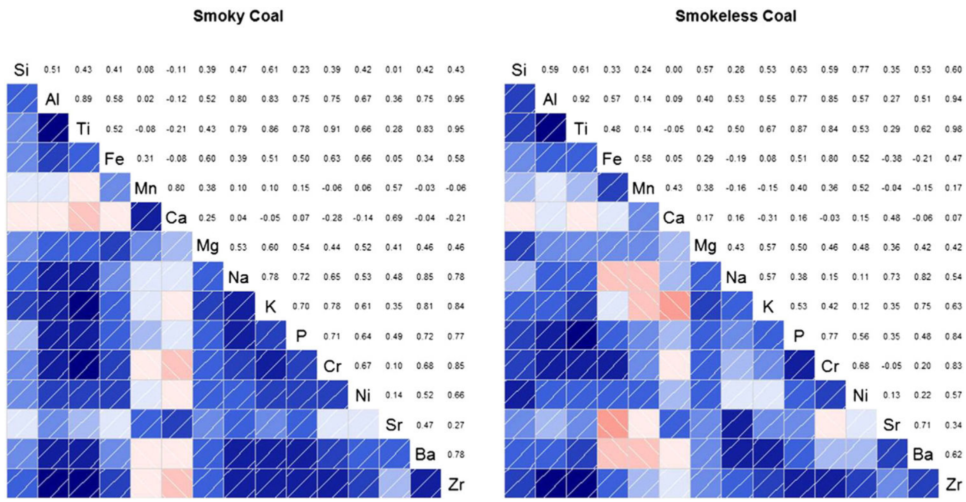

Fig. 2.

Correlogram for elemental analysis by smoky and smokeless coals. Blue colour and upward sloping lines indicate positive correlation and red and downward sloping lines indicate negative correlations. Colour intensity indicates strength of correlation.

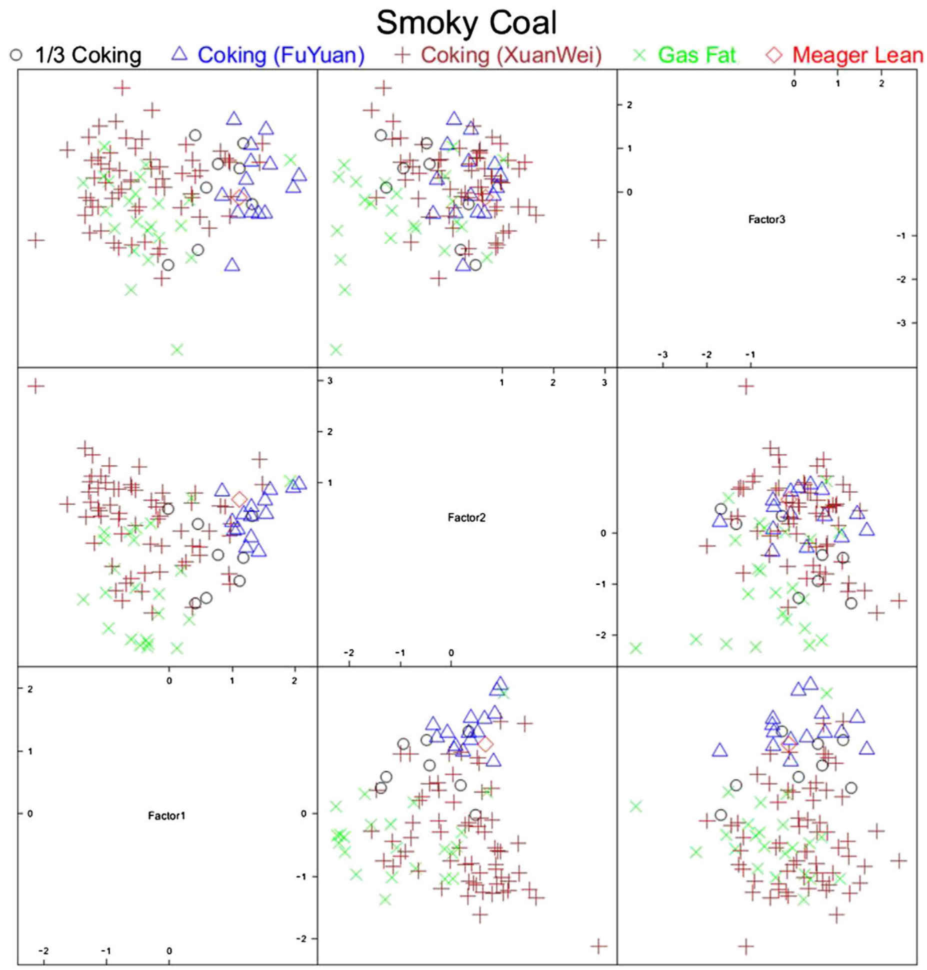

Fig. 3.
Scatter plot matrix of smoky coal factors (n = 3) by smoky coal subtype.

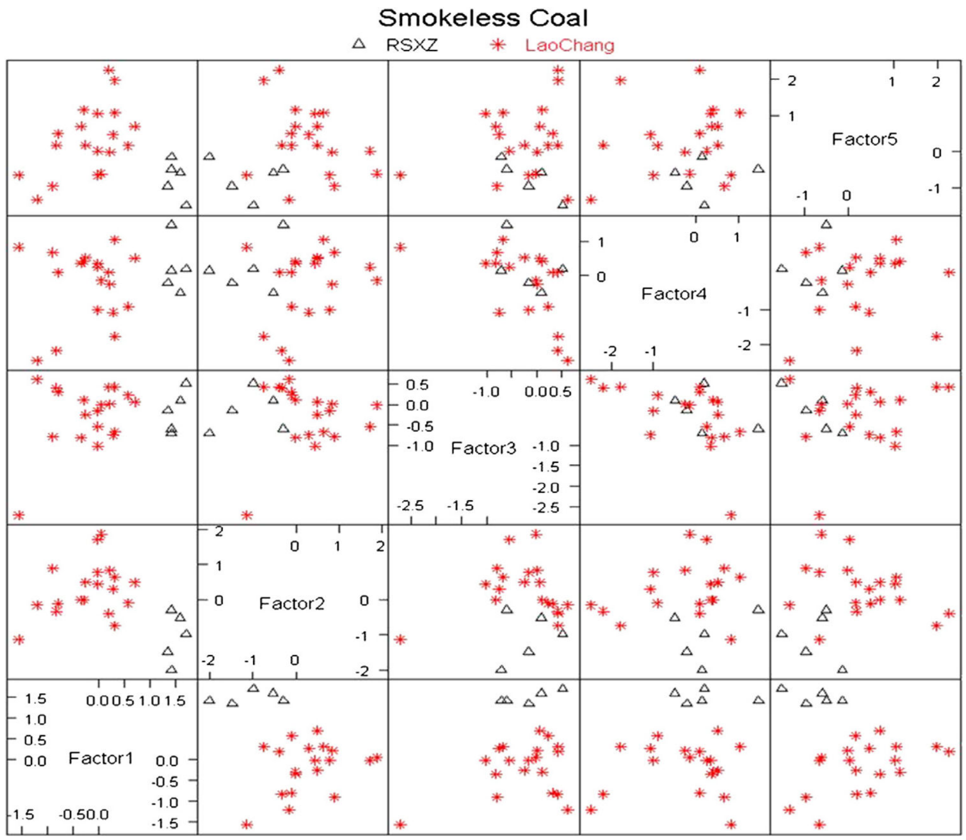

Fig. 4.
Scatter plot matrix of smokeless coal factors (n = 5) by coal producing region.

Table 1

Characteristics of smoky coal subtypes (Chen, 2000).

Coal sub-type	V_{daf}(%)	Caking index	Maximum thickness of plastic layer (mm)	Audibert-Arnu dilatation (%)
Coking coal	17.87	73	10	−1
1/3 coking coal	30.16	75	13	22
Gas-fat coal	44.27	100	36	246
Meagre lean coal	16.14	16	4	N/A

V_{daf}: dry ash free volatile matter.

Maximum thickness of plastic layer: a representation of coal rank and petrographic composition.

Audibert-Arnu dilatation: a measure of expansion and contraction characteristics of coal.

Table 2

Average carbon, sulfur and trace element levels for coals from USA and China (Bragg et al., 1998; Dai etal., 2008b).

	USA coals	China coals
Total carbon (%)	70	-
Sulfur (%)	2.2	-
Si	23,900	39,700
Al	14,110	32,000
Ti	796	2218
Fe	15,103	40,400
Mn	33	124
Ca	2637	10,000
Mg	632	1500
Na	377	1320
K	1847	1742
P	203	410
Cr	16	15
Ni	17	14
Sr	109	140
Ba	101	159
Zr	23	89
Zn	72	42
Pb	11	15
Co	7	7
Cu	17	18

Unless specified otherwise, values are in ppm.

Table 3

Geochemical analysis, carbon and sulfur contents and ash yield, stratified by fuel type and source (median values).

Major coal type	County	Coal sub-type	Coal mine	N	S1 (mg HC/g coal)	S2 (mg HC/g coal)	Tmax (°C)	Total carbon (%)	Inorganic carbon (%)	Organic carbon (%)	Total sulfur (%)	Ash yield (%)
Smoky	Xuanwei	Coking coal		**116**	**2.15** ^	**71. 5** ^	**460** ^	**59.39**	**0.5**	**58.81**	**0.24** ^	**32**
				_65_	_2.35_	_73.24_	_462_	_60.76_	_0.53_	_60.46_	_0.2_	_32_
				63	2.34 *	73.24 *	462 *	60.76 *	0.52 *	60.46 *	0.2	33
			Azhi	15	2.52	75.62	462	65.53	0.92	64.6	0.23	30
			Baoshan	5	2.24	61.59	463	57.3	0.64	54.17	0.18	37
			Laibin	14	2.23	76.14	459.5	55.76	0.57	54.97	0.28	35
			Tangtang	22	1.62	61.98	464	55.7	0.19	55.22	0.18	38
			Yangchang	7	4.03	138.69	443	70.12	0.32	69.77	0.19	23
	Fuyuan	Coking coal		_51_	_1.95_	_68.86_ +	_452_ +	_57.28_ +	_0.43_ +	_55.79_ +	_0.34_	_29_ +
				15	1.89	43.96	467	50.83	1.8	46.42	0.24	36
			Daping	4	2.34	33.9	458.5	43.58	0.8	42.74	0.29	40
			Enhong	4	1.73	40.53	469.5	50.23	5.56	42.26	0.74	51
			Haidan	6	1.78	46.75	468	46.29	3.01	43.28	0.19	32
			Zude	1	4.21	61.02	473	64.15	0.4	63.75	0.87	23
		1/3 coking		8	1.98	52.35	459 *	54.33	0.34	53.67	0.36 *	29
			Bagong	5	1.7	47.19	465	53.32	0.13	52.14	1.88	29
			Dahe	3	3.08	80.18	445	65.46	0.43	64.89	0.16	25
		Gas fat		23	1.97	131.87	433	64.34	0.2	63.7	0.39	24
			Housuo	20	2.03	138.44	433.5	63.98	0.24	63.6	0.4	25
			Qingyun	3	1.62	110.4	431	65.88	0.12	65.83	0.29	29
		Meagre lean	Gumu	1	3.83	53.43	469	54.7	1.4	53.3	0.53	47
Smokeless				**29**	**0.29**	**8.39** *	**581**	**57.41** *	**0.3**	**56.92** *	**1.03** *	**32**
	Fuyuan		LaoChang	_19_	_0.28_	_9.47_	_571_	_57.41_	_0.22_	_57.33_	_1.03_	_31_
	Xuanwei		RSXZ	_5_	_0.29_	_6.49_	_572_	_46.17_	_0.15_	_45.81_	_2.83_	_41_

S1: mg of hydrocarbons (per gram of coal) released under controlled burning at 300 °C.

S2: mg of hydrocarbons (per gram of coal) released during maximal release of hydrocarbons under controlled burning.

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Tmax: temperature at which S2 occurs.

Italicized values indicate overall value for each county.

^ Statistically significant variation between smoky & smokeless coal types.

† Significant variation within smoky coal subtypes for that region.

* Significant variation between mines within that coal type in that region.

Table 4

Elemental composition (in parts per million) of coal samples, stratified by fuel type and source (median values).

Major coal type	County	Coal sub-type	Coal mine	N	Si	Al	Ti	Fe	Mn	Ca	Mg	Na	K	P	Cr	Ni	Sr	Ba	Zr
Smoky		Coking coal		**116**	**92,100**	**19,750** ^	**1400** ^	**18,400**	**200**	**8400**	**1700** ^	**200** ^	**700** ^	**100** ^	**20** ^	**29**	**53**	**37** ^	**66** ^
	Xuanwei			*65*	*102,100*	*16,600*	*1000*	*19,100*	*500*	*14,400*	*1800*	*200*	*500*	*100*	*15*	*29*	*45*	*28*	*45*
				63	102,300 *	16,900	1000	19,100 *	400 *	13,000 *	1800 *	200 *	600	100	15 *	29 *	45 *	29	45
			Azhi	15	80,800	13,000	600	10,300	700	27,900	1500	200	400	100	8	22	83	25	32
			Baoshan	5	99,100	25,300	1100	14,000	400	12,800	1400	200	500	100	12	25	39	25	93
			Laibin	14	115,050	16,200	850	27,700	600	17,900	1850	200	400	100	14	33	42	28	41
			Tangtang	22	115,750	22,500	1750	27,800	200	3950	2050	250	1100	100	32	34	31	35	71
			Yangchang	7	72,800	13,200	1000	6400	200	7200	1000	200	400	100	11	18	80	38	37
	Fuyuan			*51*	*85,100*	*30,400* +	*3800* +	*18,100* +	*200* +	*5500* +	*1400* +	*300* +	*1700* +	*300* +	*37* +	*29* +	*59* +	*95* +	*141* +
		Coking coal		15	121,100	49,700	7400	30,200	300	8600	3200	1100*	3900	600	50	43	94	240	200
			Daping	4	112,100	54,050	7950	34,750	250	9550	2850	750	4900	850	64	39	95	215	218
			Enhong	4	97,650	41,650	6700	27,900	300	10,750	5150	850	2950	450	60	35	108	207	191
			Haidan	6	121,150	55,000	10,500	30,850	400	5100	3000	1350	5100	750	60	48	92	418	276
			Zude	1	57,100	40,500	5000	16,300	100	8600	2500	400	3600	300	41	30	97	88	152
		1/3 coking		8	79,750	41,550	5850	27,900 *	200	3650 *	800	300	1700	250	60 *	27	52 *	95	208
			Bagong	5	79,800	43,900	6500	36,100	200	1000	600	300	1700	300	63	26	36	95	238
			Dahe	3	79,700	22,000	1700	5900	200	8200	900	200	1200	200	14	28	83	95	73
		Gas Fat		23	77,100	14,900	900	12,500	0	2400 *	1000	200	200	100	13	22	16 *	29	46
			Housuo	20	79,000	13,600	950	12,200	0	2600	850	200	250	100	13	23	24	28	45
			Qingyun	3	54,200	18,100	900	19,600	0	400	1200	200	200	100	12	22	10	33	50
		Meagre lean	Gumu	1	87,400	51,000	5400	19,400	300	18,900	3900	500	4900	400	41	28	99	115	174
Smokeless				**29**	**84,000**	**43,800** *	**5100** *	**22,100** *	**200**	**7800**	**2000** *	**600**	**4100** *	**300**	**35** *	**25** *	**69**	**109** *	**157** *
	Fuyuan		*LaoChang*	*19*	*85,700*	*36,800*	*4700*	*17,700*	*200*	*7800*	*2500*	*700*	*4900*	*300*	*29*	*22*	*74*	*130*	*148*

Major coal type	County	Coal sub-type	Coal mine	N	Si	Al	Ti	Fe	Mn	Ca	Mg	Na	K	P	Cr	Ni	Sr	Ba	Zr
	Xuanwei		*RSXZ*	*5*	*77,900*	*71,000*	*7500*	*47,500*	*200*	*6000*	*1400*	*300*	*2500*	*300*	*75*	*31*	*19*	*65*	*223*

Italicized data indicate overall value for each county.

^ Statistically significant variation between smoky & smokeless coal types.

+ Statistically significant variation within that smoky coal subtypes for that region.

* Statistically significant variation between mines within that coal type in that region.

Table 5

Findings of exploratory factor analysis on elemental composition.

Smoky coal				Smokeless coal				
Trace elements	**Factor 1**	**Factor 2**	**Factor 3**	**Factor 1**	**Factor 2**	**Factor 3**	**Factor 4**	**Factor 5**
EVs (% variance explained)	6.6 (44%)	2.5 (17%)	2.4 (16%)	5.1 (34%)	3.4 (22%)	1.8 (12%)	1.6 (10%)	1.3 (8%)
Si	0.37	−0.03	0.36	0.39	0.30	**0.65**	−0.06	0.33
Al	**0.86**	0.02	0.37	**0.89**	0.25	0.20	0.05	0.08
Ti	**0.92**	−0.09	0.30	**0.90**	0.34	0.17	−0.10	0.10
Fe	0.29	0.05	**0.90**	**0.68**	−0.52	0.21	0.13	0.25
Mn	−0.12	**0.86**	0.33	0.19	−0.26	0.40	0.47	0.27
Ca	−0.11	**0.93**	−0.09	0.00	0.16	0.02	**0.98**	0.00
Mg	0.38	0.37	**0.50**	0.24	0.26	0.33	0.12	**0.78**
Na	**0.84**	0.18	0.16	0.27	**0.81**	−0.02	0.04	0.13
K	**0.83**	0.08	0.33	0.46	0.54	−0.07	−0.40	**0.50**
P	**0.77**	0.20	0.29	**0.78**	0.25	0.22	0.12	0.23
Cr	**0.76**	−0.17	0.47	**0.89**	−0.14	0.32	−0.02	0.22
Ni	**0.53**	−0.03	**0.55**	0.43	−0.01	**0.89**	0.13	0.09
Sr	0.43	**0.79**	−0.14	0.02	0.84	0.09	0.35	0.09
Ba	**0.88**	0.09	0.07	0.29	**0.89**	0.13	−0.20	0.14
Zr	**0.90**	−0.09	0.36	**0.90**	0.37	0.21	0.00	0.06

Factor: latent variable comprising of multiple correlated variables which assists in explaining overall variance in dataset.

Variables in bold are considered to be contributory to that factor.

Factor analysis performed under varimax rotation.

Table 6

Total and respirable quartz levels as measured from scanning electron microscopy (SEM).

Major coal type	**County**	**Coal sub-type**	**Coal mine**	**N**	**Total Quartz (% of coal)**	**Respirable Quartz*** **(% ofcoal)**
Smoky	Xuanwei	Coking		**19**	**4.58**^	**1.93**^
				11	*6.77*	*3.1*
				10	7.52	3.39
			Azhi	2	4.11	2.33
			Baoshan	1	6.4	3.83
			Laibin	2	10.48	5.69
			Tangtang	4	7.52	3.39
			Yangchang	1	11.97	1.28
	Fuyuan	Coking		*8*	*3.24*	*1.14*
				3	4.58	1.87
			Daping	1	3.82	0.93
			Enhong	1	7.39	2.46
			Haidan	1	4.58	1.87
		1/3 coking		2	1.87	0.98
			Bagong	1	1.08	0.63
			Dahe	1	2.66	1.33
		Gas fat	Housuo	2	16.5	9.83
		Meagre lean	Gumu	1	2	0.96
Smokeless				**6**	**2.24**	**0.60**
	Fuyuan		LaoChang	*4*	*2.41*	*0.85*
	Xuanwei		RSXZ	*2*	*1.77*	*0.27*

Italicized data indicate overall value for each county.

^Statistically significant variation between smoky & smokeless coal types.

*Respirable quartz represents quartz grains of size less than 9.6 μm.