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Geological and geochemical characteristics of high arsenic coals from endemic arsenosis areas in southwestern Guizhou Province, China

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Abstract

Southwest Guizhou Province is one of the most important areas of disseminated, sediment-hosted-type Au deposits in China and is an important area of coal production. The chemistry of most of the coals in SW Guizhou is similar to those in other parts of China. Their As content is near the Chinese coal average, but some local, small coal mines contain high As coals. The highest As content is up to 3.5 wt.% in the coal. The use of high As coals has caused in excess of 3000 cases of As poisoning in several villages. The high As coals are in the Longtan formation, which is an alternating marine facies and terrestrial facies. The coals are distributed on both sides of faults that parallel the regional anticlinal axis. The As content of coal is higher closer to the fault plane. The As content of coal changes greatly in different coal beds and different locations of the same bed. Geological structures such as anticlines, faults and sedimentary strata control the distribution of high As coals. Small Au deposits as well as Sb, Hg, and Th mineralization, are found near the high As coals. Although some As-bearing minerals such as pyrite, arsenopyrite, realgar (?), As-bearing sulfate, As-bearing clays, and phosphate are found in the high As coals, their contents cannot account for the abundance of As in some coals. Analysis of the coal indicates that As mainly exists in the form of As^{5+} and As^{3+}, perhaps, combined with organic compounds. The occurrence of such exceptionally high As contents in coal and the fact that the As is dominantly organically associated are unique observations.

1. Introduction

Historically, As has been used in medicine, agriculture, and industry. Although there is evidence that As is an essential element for animals there is no consensus that it is essential for humans (North et al., 1997). Arsenic can be one of the most hazardous elements known, although its acute lethal dose is unknown (North et al., 1997).

Endemic As poisoning in China is widespread and locally very severe. In China, endemic As poisoning is known to be caused by a variety of processes including the use of As-bearing underground water and As-rich coal.

Endemic As poisoning caused by domestic coal combustion has been found in many places in China since the 1970s (Ke et al., 1980; Sun et al., 1984; Zhou et al. 1993; Zhou et al., 1994). In the countryside of China many people rely on domestic coal combustion for cooking and heating where the coal is commonly burned in unventilated stoves. It is common practice in the humid mountainous areas of south China for peasants to dry their corn and other vegetables directly over the coal

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embers. This practice has led to serious endemic arsenosis, especially when mineralized coals are used.

The most serious endemic arsenosis caused by domestic coal combustion occurs in SW Guizhou Province where the local people use high As coals. In this area, the arsenosis is widespread and the incidence of endemic As poisoning is very high. More than 3000 patients in the mountainous region of SW Guizhou Province have been diagnosed with arsenosis.

There have been many studies on the high As coals from south China. Long et al. (1993) suggests that the distribution of high As coals was controlled by sedimentary strata and geological structures, and that there is a relation to mineralization of Au, Sb, and other elements. Zheng et al. (1994, 1996, 1997, 1999) studied the factors that control high As coals and the ways in which As entered the local environment and human bodies. Belkin et al. (1997a,b) reported preliminary chemical and mineralogical results on the high As coals from this area. Zhao et al. (1998) studied these coals using extended X-ray fine structure (EXAFS) spectroscopy. Ding et al. (1999) studied the mode of occurrence of As in these coals.

The authors have conducted field geology and laboratory examination of the coals by low temperature ashing (LTA) and X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy-energy dispersive X-ray analysis (SEM-EDX), EXAFS, instrumental neutron activation analysis (INAA), and wet chemical analysis. This paper describes these extraordinary As-rich coals and summarizes their distribution and controlling factors, including the sedimentary facies of high As coals, the mode of occurrence of As, and the processes of As concentration in the coals.

2. Geology

Southwest Guizhou (Fig. 1) is located near the edge of a Precambrian craton, 1 of 3 Precambrian cratons in China. Most of Guizhou Province is underlain by the Yangtze craton, which is composed of Proterozoic crystalline rocks overlain by Paleozoic and Lower to Middle Triassic marine deposits and Upper Triassic terrestrial deposits (Bureau of Geology and Mineral Resources of Guizhou Province, 1986).

Devonian to Triassic shallow-marine carbonate rocks were deposited on a broad cratonic platform near the edge of the Yangtze craton. The area was uplifted during late Triassic time and was covered by widespread terrestrial deposits, including precursors to the coal beds.

The rocks were pervasively folded and faulted during the Yanshanian orogeny, 190–65 Ma. ago. Gently to locally moderately tight folds exist, sometimes associated with high-angle faults. Most of the rocks at the surface in SW Guizhou Province are Triassic in age. Permian rocks generally crop out in the cores of anticlines and in partly exposed reefs that were topographic highs during the Triassic (Bureau of Geology and Mineral Resources of Guizhou Province, 1986). No igneous rocks are exposed in the immediate vicinity of the study area.

Coal beds occur in the Late Permian Longtan formation, which is an alternating marine facies and terrestrial facies. The coals from large coal mines are similar to

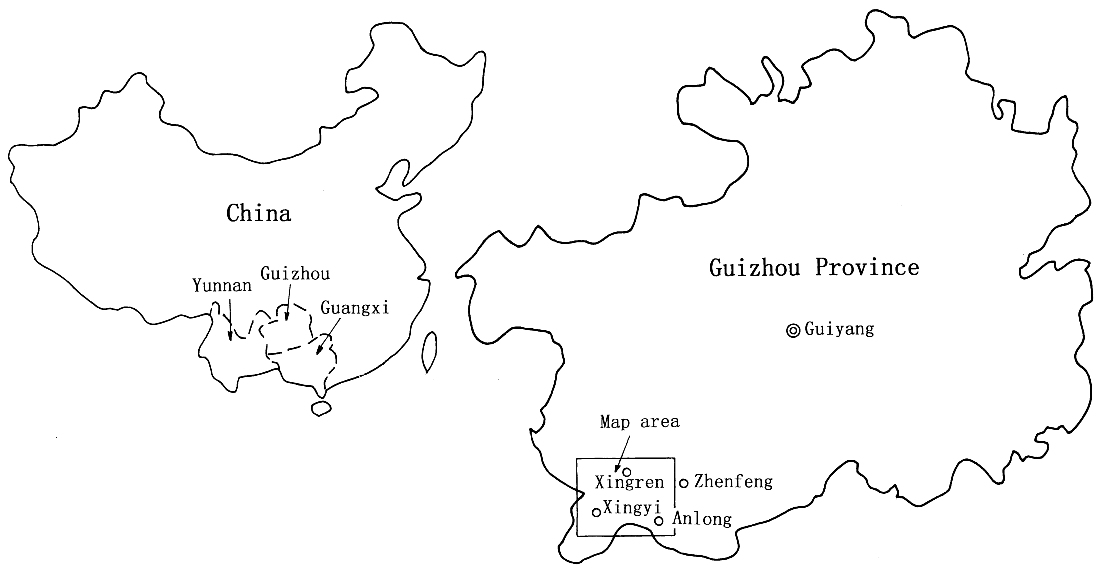

Fig. 1. Location of study area.

those elsewhere in China in that their As content is near the Chinese coal average (about 10–20 ppm), but some small, local coal mines contain high As coals. The high As coals are anthracitic, apparently due to local thermal metamorphism related to mineralization.

High As coals are located in Xingyi, Xingren, and Anlong counties which are part of the Yunnan-Guizhou-Guangxi sediment-hosted Au area (Fig. 2). Small Au deposits are found near the high As coals. Lenticular ore bodies of orpiment, which were linked by veinlets, were found in a small Au deposit near samples with the highest As contents. Strata in the study area are limestone of the Lower Permian Maokou formation, clay rock and sandstone mixed with coals and limestone of the Late Permian Longtan formation, flinty carbonate of the Changxing formation, siliceous shale of the Dalong formation, and fine-grained sandstone of the Triassic Yelang formation.

3. Samples studied and methods of analysis

Thirty-three samples were analyzed for this study. These included 19 samples from Anlong County, 13 samples from Xingren County, and 1 sample of anthracite from Xingri County. The samples from Anlong included 13 anthracite samples (grab samples from dumps and mine samples) and 6 rock samples associated with the coal or Au deposits. The samples from Xingren included 10 anthracite samples (grab samples from dumps and mine samples) and 3 rock samples associated with the coal.

Reference chips were removed from the samples to prepare thin sections and the remainder of the material was ground to pass a 200-mesh sieve. The chips were mounted in epoxy plastic, and polished surfaces were prepared both normal and parallel to bedding. The polished thin sections were examined by both incident-light and transmitted-light microscopy. Some of the samples were

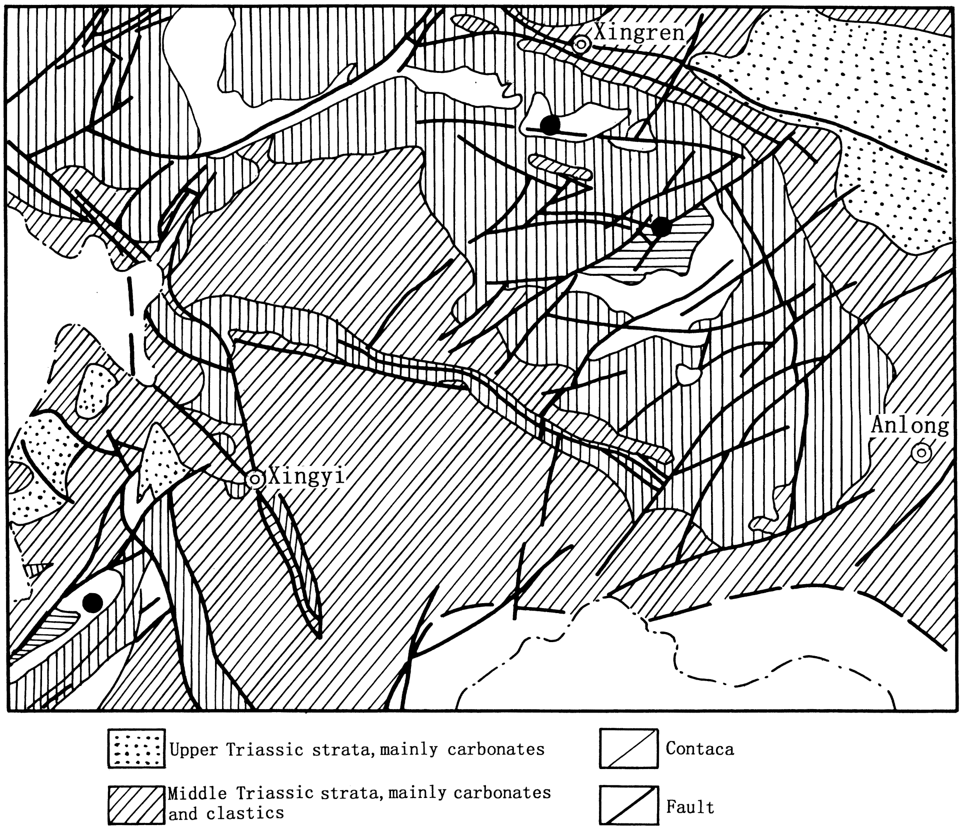

Fig. 2. Geological map of study area.

examined by an electron microprobe analyzer (EMPA) and by SEM-EDX. The ground material was split and a sub-split was subjected to low temperature ashing (LTA), and X-ray diffraction analysis of raw coal and LTA product.

Several samples were analyzed by high-energy X-rays in a synchrotron to determine the As-bearing phase. Two of the high As samples were analyzed in the USA and these 2, plus 4 other samples were analyzed at the Institute of High Energy Physics, Chinese Academy of Sciences (IHEPCAS). The two high As samples were examined by TEM in China and one was examined in the USA.

Splits of the ground material were analyzed for As and other trace elements by instrumental neutron activation analysis. Some of the samples were also analyzed by wet chemical analysis by the Chinese Standard Test Method (GB 3085-82).

4. Chemistry

The contents of Fe, Au, Sb, As and Hg in Guizhou Province coal and rock samples are listed in Table 1. Three different laboratories examined the samples with different analytical methods and obtained similar results. The concentration of As in the coals (up to 3.5 wt.%: all values are on a coal basis) in parts of Guizhou Province is much higher than the content of As in coals in other parts of China and in the USA. The As levels found in these coal samples are also higher than those of other high As coals reported in the literature (Swaine, 1990). The Hg contents (up to 45 ppm) of these coals are also extremely high.

The abundance of As correlates with that of Sb. Iron shows a wide range of values from 0.18 to 7.75%, with no apparent correlation with As, chalcophile elements, or locality. Although limited by a small data set, these data indicate that As and Fe do not have the same modes of occurrence in every sample.

The distribution of the high-As coals is related to rock structure and Au-mineralization. Samples Ah-1-1–Ah-1-4 are rocks from a Au deposit. The concentration of As in the brecciated samples (600–800 ppm) is much higher than the As in the associated carbonaceous shale (200 ppm). The authors infer that the As concentrations were controlled by the greater permeability of the breciated rock.

The concentration of As is very uneven, even in the same location or in the same coal seam. Samples Xn-4-1 (400–500 ppm) and Xn-4-2 (1600 ppm) were taken from the same mine location but a nearby sample (Xn-4-3) had 3400 ppm. Samples Ah-3-1 (860 ppm), Ah-3-2 (570 ppm) and Ah-3-3 (280 ppm) were taken from the same bed. Samples Xn-e, Xn-3-1 and Xn-3-2 were collected from different places in the same bed. The As content of these samples ranges from 1200 to 7900 ppm. The associated rocks (Xn-3-3, Xn-3-4, and Xn-3-6) have As contents of 150–590 ppm. Liu et al. (1989) found that coal samples near the Au deposit had an average As content of 270 ppm with many values of more than 1000 ppm and an average Au value of 54.5 ppb. The maximum Sb content they found was 600 ppm with an average of 67 ppm.

Three coal samples [Ah-2-1 (32,000 ppm), Ah-2-2 (35,000ppm), and Ah-2-3 (34,000 ppm)] collected near a small abandoned adit may have the highest As concentrations ever recorded from a coal. Sample Ah-2-2 was taken just inside, and Ah-2-3 was taken just outside this portal. Sample Ah-2-1 was collected, as a grab sample, from the rubble at the portal entrance. Samples Ah-2-10 (320 ppm) and Ah-2-11 (200 ppm) were collected about 50 and 10 m, respectively, from the portal. The associated rock samples, Ah-2-7 (74 ppm), Ah-2-8 (110 ppm), and Ah-2-9 (94 ppm) had relatively high As contents for this region.

5. Arsenic distribution

Sedimentary strata and structures such as folds and faults controlled the distribution of the high As coals. High As coals are distributed on both sides of the faults that parallel the regional anticlinal axis. The As content of the coal is higher the closer the coal is to the fault plane (Fig. 3).

Small deposits of Au and/or Sb, As, and Hg were found in sedimentary rocks near the high As coals. The concentrations of As, Sb, Au and Hg are high in both the high As coals and the Au deposits, indicating a genetic link. Fleet and Mumin (1997) thought that sulfides containing Au formed solid solutions with sulfides with high As and low Fe. He and Wu (1997) suggested that a deep fluid with Au, As and other elements rose along faults and extracted ore-forming elements from strata, separating under suitable conditions to form different deposits.

The range of high As coals may be larger than what is known. Zhou and Ren (1992) found high As coals in east Yunnan Province, which is adjacent to SW Guizhou and also in part of the Yunnan-Guizhou-Guangxi Au impregnated area. Cunningham et al. (1988) found high As coals in Zhenfeng county, Guizhou Province. But arsenosis was not found in either east Yunnan or Zhenfeng.

6. Geochemistry of high arsenic coals

Knowing the form or mode of occurrence of an element in coal can be very useful. To anticipate the environmental impact, technological behavior, or by-product potential of an element requires information on its mode of occurrence (Finkelman, 1981, 1995). It is commonly believed that As in coal is primarily found with the sulfide minerals such as pyrite (Bouska, 1981; Finkelman, 1981, 1995; Coleman and Bragg, 1990; Swaine, 1990; Hower et al., 1997). Palmer et al. (1993) studied the modes of occurrence of As in US coals and concluded that it was mostly associated with pyrite, organically-bound As was a minor constituent, and arsenopyrite was a trivial phase. Huggins and Huffman. (1996a,b) and Huggins et al. (1996) found that much As existed in the form of $[AsO_4]^{3-}$ in the coals that they studied. But they thought that the $[AsO_4]^{3-}$ resulted from the oxidation of As-bearing pyrite.

Table 1
Contents of As, Sb, Au and Fe in coal and rock samples from SW Guizhou Province. All values are on a coal basis

Method	INAA[d]	INAA	INAA	GB[e]	INAA	GF[f]
Laboratory	USGS	USGS	USGS	IGCAS	USGS	USGS
Sample	Fe%	Au (ppb)	Sb (ppm)	As (ppm)		Hg (ppm)
Xn-102[a]	0.84		73		1700	15
Xn-103	0.84		55		2200	14
Xn-e	4.63	350	170		7900	8.5
Xn-5	0.18	190	40		600	29
Xn-10	7.75	28	28		400	2.0
Xn-4-1	6.2	570	130		1600	45
Xn-4-2				1600		
Xn-4-3				3400		
Xn-3-1				1400		
Xn-3-2				2100		
Xn-3-4 (sandstone)				520		
Xn-3-5 (pyrite)				150		
Xn-3-6 (clay)				590		
Ah-2-1[b]	0.25	<1	140		32000	5.8
Ah-2-2	0.34		210		35000	4.1
Ah-2-3	0.64		210		34000	3.2
Ah-2-10	0.5	3.6	8.0	53	48	0.32
Ah-2-11	3.5	6.1	13	290	320	0.48
Ah-9	1.3	5.8	24		200	2.2
Ah-10	2.2	7.1	9.3	120	120	0.64
Ah-11				5.3		
Ah-1-1 (breccia)				610		
Ah-1-2 (breccia)				760		
Ah-1-3 (breccia)				850		
Ah-1-4 (shale)				190		
Ah-3-1				860		
Ah-3-2				570		
Ah-3-33				280		
Ah-LY				470		
Ah-2-7 (pyritic shale)				74		
Ah-2-8				110		
Ah-2-9				94		
Xi-1[c]	0.64	6.4	1.7	1100	1100	0.26

[a] Xn = Xingren County.
[b] Ah = Anlong County.
[c] Xi = Xingyi County.
[d] INAA = instrumental neutron activation analyses.
[e] GB = Chinese Standard Test Method GB 3085-82.
[f] GF = graphite furnace atomic absorption spectroscopy.

Only a few workers have suggested that As in coal may be in an organic form. Mechàcek and Petríck (1967) suggested that As-bearing organic compounds (unnamed) coexist with As-bearing pyrite, orpiment, realgar.

The main As-bearing minerals in Guizhou Province coals are pyrite and minor arsenopyrite. The gangue minerals are quartz, calcite, and some clay minerals. Arsenopyrite was found to co-exist with As-bearing pyrite. Quantitative electron microprobe analyses indicate that there are large differences in the As content in pyrites formed during different periods. Pyrite formed during the peat stage is common but its As content is low, whereas pyrite formed during the period of Au mineralization has high As. The EMPA data in Table 2 illustrates the lower As in pyrite cores compared to overgrowths. Usually the abundance of As in pyrite is not more that 3 wt.% (Wang et al., 1984). A high content of As in vitrinite was also determined by EMPA.

X-ray diffraction studies of raw coal and LTA were conducted to identify the As-bearing species in the high As samples. The XRD results indicate that a wide variety of minerals are present, however, no specific As minerals were identified.

Samples were examined by X-ray absorption fine structure (XAFS) with a synchrotron at the IHEPCAS and at the University of Kentucky. Collection of spectrum intensities across the XANES (X-ray absorption near-edge structure) and EXAFS (extended X-ray absorption fine structure) regions of an absorption spectrum can provide information on the electronic state and chemical coordination for each crystallographic site of the chosen element. Results from the IHEPCAS demonstrated that, for most samples, the nearest atom to As is O, with a coordination number of 4. Results from the University of Kentucky demonstrated that, in one high

Table 2
Composition of pyrite in sample Xn-e (electron microprobe analyses in wt.%)

Grain		As	Au	Fe	S	Sb	Cu	Hg	Sn	Sum
L2742	Core	1.03	0.24	47.00	51.72	0.04	0.02	–	–	100.05
	Rim	4.50	0.34	45.79	49.75	0.09	0.04	0.02	0.01	100.60
L2361	Core	2.75	0.36	46.05	51.47	0.09	0.02	0.14	0.01	100.86
	Rim	5.76	0.37	43.39	47.27	0.12	0.12	0.47	0.07	97.53

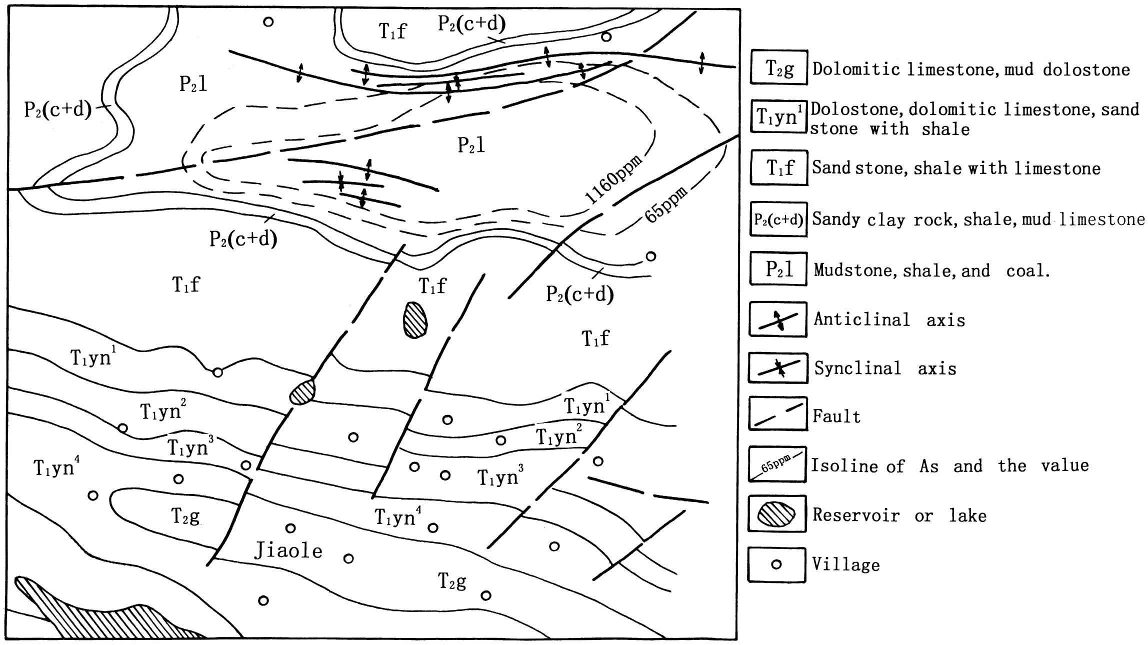

Fig. 3. The distributed regularity of high As-bearing coals in Jialoe, Xingren County (modified from Zhou et al., 1991).

As sample 100% of the As occurred as $[AsO_4]^{3-}$ and in another sample about 75% of the As occurred as $[AsO_4]^{3-}$ with the balance as sulfide bound As. Thus, for the coals examined, the preponderance of the As is mainly in the 5+ state valance state.

The samples with the highest As concentrations (those greater than 3 wt.%) are from Anlong County and are mineralogically unusual. Although they contain small grains and veins of As-bearing sulfate, As-bearing pyrite, and As-bearing clay, these phases are completely inadequate to account for the As abundance. In the SEM many of the coal samples exhibit a box-like arrangement of a filmy white material (Fig. 4). The bands range from a few to tens and hundreds of microns in thickness. Semi-quantitative analysis by SEM-EDS on many randomly selected areas demonstrated that the bright bands contain As at levels >3 wt.%. Iron is low but always present at levels from 0.2 to 0.4 wt%. Sulfur and C are the only other major elements observed. The SEM (50,000x) did not detect any discrete As-bearing phases is these bands. A fragment of one sample was examined by TEM. No discrete As-bearing phase could be observed using this instrument at magnifications of 1 million times. So the possibility of finely-dispersed arsenopyrite or As-bearing pyrite as the origin of the As can be ruled out.

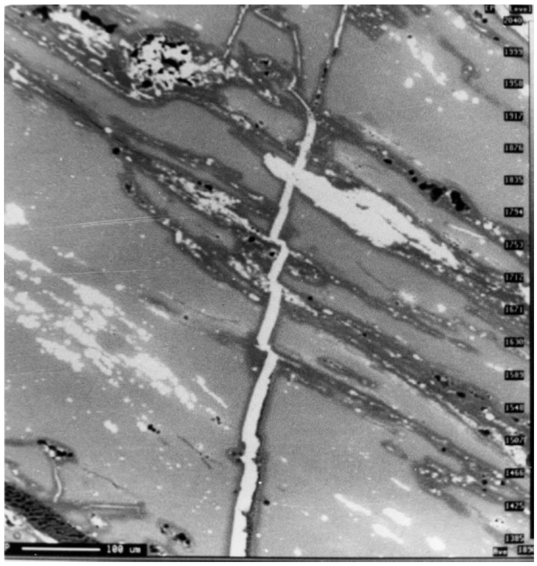

Fig. 4. Back-scattered scanning photomicrograph of a polished block of As-rich coal from Anlong County, China. Dark areas are coals, bright areas are minerals (mainly pyrite), light grey areas are organically-bound As. Scale bar is 100 μm.

7. Emplacement of arsenic

The abundance of As in some Guizhou Province coal samples is more than 3 wt.%. The concentration of As in most terrestrial plants is generally not more than 1 ppm. It is highly unlikely that plants would be able to concentrate As at levels high enough to account for the As in the Guizhou Province coals. Arsenic could be introduced during peat formation and/or during later stages in the coalification process. Abundant pyrite formed during the peat stage, but its content of As is low. Pyrite formed during later stage mineralization has high As contents but is not as common.

The textural evidence, arsenate saturating the organic matter, indicates that As in parts of Anlong County was emplaced early, perhaps during peatification. In other parts of the Guizhou Province, the As is primarily in fracture-filling pyrite and arsenopyrite and was deposited by mineralizing solutions later in the coalification process.

8. Concluding comments

Coals in southwestern Guizhou Province, PR China have exceptionally high contents of As, Hg, Sb and Au. Mineralization of the coals by hydrothermal fluids probably took place at several stages from the peat-forming period to late-stage epigenesis. Coals in Anlong County contain as much as 3.5 wt.% As on a coal basis. Most of the As occurs as arsenate ions in the organic matrix. Use of the mineralized coal for domestic energy needs has resulted in more than 3000 cases of severe As poisoning. Additional work is needed to better understand: the distribution of high As coals in the region; the forms of As in the coal; the relationship of As content to that of Au, Sb and Hg; the behavior of As during combustion; the reaction of volatilized As with foods being dried over the coal-burning ovens; and the human health impacts of multiple pathway exposure to high levels of As and other potentially toxic elements.

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