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Method for evaluation of the cleanliness grade of coal resources in the Huainan Coalfield, Anhui, China: a case study

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Abstract Based on analysis of a large data set and supplementary sampling and analysis for hazardous trace elements in coal samples from the Huainan Coalfield, a generalized contrast-weighted scale index method was used to establish a model to evaluate the grade of coal cleanliness and its regional distribution in the main coal seam (No. 13-1) The results showed that: (1) The contents of Cr, Mn and Ni in the coal seam are relatively high and the average values are greater than 20 μg/g. The contents of Se and Hg are at a high level while most other trace elements are at normal levels. (2) The cleanliness grade of the coal seam is mainly grade III–IV, which corresponds to a relatively good-medium coal cleanliness grade. However, some parts of the seam are at grade V (relatively poor coal cleanliness). (3) Coal of relatively good cleanliness grade (grade III) is distributed mainly in the regions corresponding to the Zhuji-Dingji-Gubei coal mines and in the eastern periphery of the Panji coal mine. Coal of medium cleanliness (grade IV) is distributed mainly in the regions of the Panji-Xiejiaji and Kouzidong coalmines. Relatively poor grade coal (grade V) is distributed in the southwest regions of the coalfield and the contents of Cr, As and Hg in coal collected from the relatively poor coal cleanliness regions often exceed the regulatory standards for the maximum concentration limits.

Keywords Coal cleanliness · Hazardous trace elements · Evaluation method · Huainan coalfield

1 Introduction

According to the yearbook of China's National Bureau of Statistics, by the end of 2017, coal accounted for 60.4% of China's primary energy consumption structure, thus a change of the coal-dominated energy structure to greener sources of energy in the short-term would be challenging. Research concerning the evaluation of coal resources in terms of their coal cleanliness grade has important implications for environmental, social and economic significance (Tang et al. 2006; Yang et al. 2011). According to the "China Clean Coal Geological Research" (2006), the evaluation index system for grading coal resources in terms of the cleanliness grade contains four elements, namely, the coal category, the toxicity, the migration behavior and the environmental protection requirements with respect to hazardous trace elements. However, due to the limitations in existing test data, the selection of evaluation indexes and the evaluation methods for establishing the cleanliness grade of coal resources in different regions of China is not the same. Up to now, the criteria for evaluating the cleanliness grade of coal resources are not unified and are essentially still at an the exploratory stage.

Coal contains many potentially toxic trace elements. According to numerous reports including the U.S. National Resources Commission (1980), the U.S. Congress Clean

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Air Act Amendment (1990), Clarke and Sloss (1992), Finkelman (1995), Swaine (1990, 1995, 2000), Zhao et al. (1997) and Ren et al. (1999), trace elements in coal have been classified as hazardous according to the degree of toxicity. Although many formal definitions of "hazardous" have been issued, most of them refer to potentially toxic elements such as Cr, Mn, Co, Ni, As, Se, Cd, Sb, Hg and Pb (Feng et al. 2019). Tang et al. (2005) selected A_d (ash content of coal) and the following elements, S, F, Cl, Cr, Mn, As, Se, Hg, Cd and Pb, as parameters for using as indices for evaluation of the coal cleanliness grade when screening coal resources for the presence of hazardous trace elements. Thereafter, Tang et al. (2006) summarized methods for selection of the evaluation indices and proposed the selection principles for the evaluation parameters. Limited by a lack of regional research, many researchers have their own opinions on the criteria for selection of evaluation indexes for coal cleanliness, however the indices normally include A_d, $S_{t,\ d}$, F, Cl and potentially toxic trace elements in coal. For example, Yang et al. (2011) selected A_d, S, F, Cl, Cr, Mn, Co, Ni, Be, Sb, U, Mo, Th, Br, As, Se, Hg, Cd and Pb as evaluation factors for comprehensive assessment of coal collected from various regions in China. Also, Guo et al. (2018), Tao (2018), Yin et al. (2018) and He et al. (2018) have proposed their preferred evaluation indices according to the actual conditions of their respective research regions.

In the past, the evaluation indices for the cleanliness grade of coal resources in Anhui Province were mainly the A_d and the S content. The A_d and S contents in coal have been used mainly to evaluate the cleanliness of the main coal seams in the Huainan and Huaibei coalfields. However, the indices for typical hazardous trace elements in coal were not considered, including indices of the volatilization characteristics of typical trace elements in coal. Moreover, selection of the limits for potential evaluation factors were not given due consideration. Relative to other coals from China and the rest of the world, the contents of Cr, Co, Ni, Se and Pb in coal from the Huainan Coalfield coal are relatively high (Chen et al. 2011; Hu, 2019) whereas the contents of Mn and Cd are similar to those from the rest of the world. The contents of Hg, As and Sb in coal from the Huainan Coalfield are relatively low, but these elements are volatile and can readily accumulate in the air. According to Yang and Ye (2017), in terms of the regional spatial distribution, a high-low concentration zone has been established as a focal point of air pollution in Anhui Province, due mainly to the large number of coal-fired power plants in the Huainan Coalfield. Hazardous trace elements (such as Hg, As, Se, Pb, Cr, Cd, Mo, Ni and Co) can accumulate in the bottom ash and fly ash of power plants during coal combustion, which result in serious environmental pollution (Ameh 2019; Song et al. 2005; Yu et al. 2017).

In recent years, with the continuous progress in coal exploration and the increasingly prominent environmental pollution problem in Huainan, Anhui Province, a large body of research data on the coal quality resources of the province including the presence of hazardous trace elements in the coals have been acquired. Thus, it is important to establish classification standards for the grading of coal cleanliness based on current geochemical analysis methods and environmental evaluation methodologies. In this paper, comprehensive research concerning an evaluation of the cleanliness grades of coal resources in the Huainan Coalfield has been undertaken by taking into account key factors such as coal quality, and the chemical properties and concentrations of typical hazardous trace elements in the coals. For instance, the No. 13-1 coal seam, the main coal seam of the Huainan Coalfield, has been examined systematically and the environmental impact of pollution from the release of hazardous trace elements as a result of coal processing and utilization are discussed.

2 Geological setting

The Huainan Coalfield is divided into the Huainan mining area, the Panji mining area and the Fudong mining area going from east to west (Fig. 1). To date, all the coal mines in the Huainan mining area have been closed and are not operational with the exception that sampling and analytical data for hazardous trace elements in the coals are being collected. The main coal-bearing strata include the Permian Shanxi Formation, the Lower Shihezi Formation and the Upper Shihezi Formation. The total thickness of the coal-bearing strata is about 750 m, with more than 30 coal-bearing strata, and the total thickness of the coal seams is about 38 m. The mineable seams have 10–16 layers, of which the Nos. 1, 4-2, 6, 8, 11-2, 13-1 coal seams are the main minable seams. The No. 13-1 coal seam is located in the middle of the fifth coal-bearing section and is the most developed coal seam in the Carboniferous-Permian coal-bearing strata. The thickness of the No. 13-1 coal seam is 0.31–12.79 m with an average thickness of 3.70–6.02 m; most areas in the coal seam have a thickness of over 4 m. The structure of the No. 13-1 coal seam is simple-relatively simple and typically contains 1–2 layers of a dirt band. The upper part of the coal seam is about 80 m from the top of the fifth coal group and the lower part is about 65 m from the top of the No. 11-2 coal seam. The roof of the No. 13-1 coal seam consists mainly of mudstone and sandy mudstone, some siltstone and fine sandstone, while the floor is mostly mudstone and sandy mudstone.

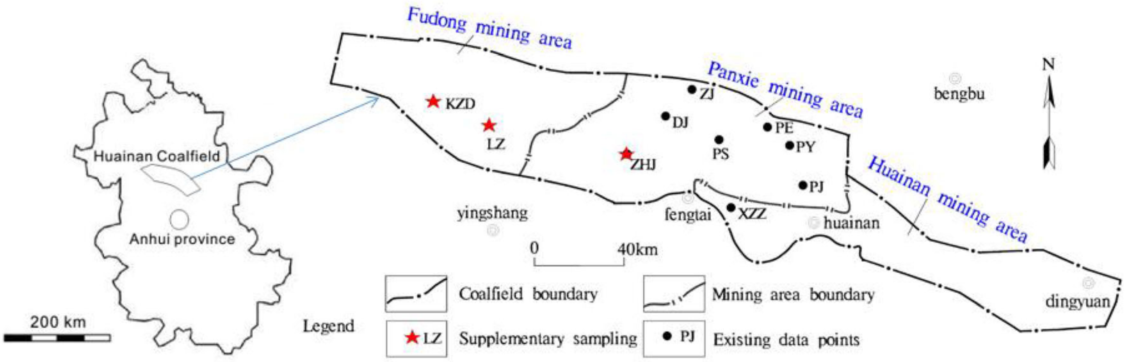


Fig. 1 Location of the Huainan Coalfield and the general situation regarding previous research

Table 1 List of coal mines and supplementary sampling points for test data (No. 13-1 coal seam)

Coalfield	Mining area	Coal mine/exploration area with existing data points	Abbreviation for coal mine	Supplementary sampling
Huainan Coalfield	Huainan mining area	Xinzhuangzi coal mine	XZZ	nd
	Panji mining area	Panyi coal mine	PY	nd
		Paner coal mine	PE	nd
		Pansan coal mine	PS	nd
		Zhuji coal mine	ZJ	nd
		Zhangji coal mine	ZHJ	Zhangji coal mine
		Dingji coal mine	DJ	nd
		Panji coal mine deep exploration area	PJ	nd
	Fudong mining area	nd	LZ	Liuzhuang coal mine
		nd	KZD	Kouzidong coal mine

Note: nd means no data

3 Sampling and methods

3.1 Data collection

Over the past 20 years, there have been ongoing studies on the presence of hazardous trace elements in coals from the Huainan Coalfield (Huang et al. 2000; Chen et al. 2009; Chen 2013; Liu et al. 2009; Kong 2007; Wu 2006; Ping 2016; Yan 2014; Chen et al. 2011, 2014; Sun et al. 2010a, 2010b; Zhou et al. 2014; Yang et al. 2012; Yan et al. 2014; Ding et al. 2018). However, the study areas have been primarily in the Panxie mining area and in the vicinity of the Huainan mining area (Fig. 1). To study and obtain a better understanding of the distribution of hazardous trace elements in the No. 13-1 coal seam, the test data for the aforementioned studies have been collated in a systematic manner, and further supplementary analyses (ZHJ, LZ, KZD samples) have been performed for the blank area (Fudong mining area).

For the survey, 313 items pertaining to the coal quality data and the trace element data (e.g., Cr, Mn, Ni, As, Se, Cd, Hg and Pb) for the No. 13-1 coal seam in eight coal mines or coal exploration areas (Table 1) in the Huainan Coalfield were collected and assessed. The data sources were mainly geological exploration reports, mining reports and published papers.

3.2 Sampling and analysis (Supplementary sampling)

In this paper, supplementary sampling was carried out in the areas where work had not previously been carried out (such as the LZ and KZD coal mines) and in some key areas where work had been carried out but the test data concerning hazardous trace elements were incomplete. There were three sampling points. The sampling horizon was the No. 13-1 coal seam of the Shangshihezi Formation. The coal samples were collected at the working faces (underground) of the coal mines. The method of slot sampling was adopted. The samples weighed more than 5 kg and were stored in sealed bags.

3.2.1 Analysis of coal quality

Proximate analysis and determination of the total S content of the three additional coal samples collected were undertaken by the Third Exploration Team of Anhui Coalfield Geology Bureau. The proximate analysis was performed according to the GB/T 212–2008. Total S analysis of coal was performed to determine the content ($S_{t,\ d}$) of dry base total S in coal and this was carried out according to GB/T–15224.

3.2.2 Determination of hazardous trace elements

Inductively coupled plasma mass spectrometry (ICP-MS) analysis of coal was performed in the Physical and Chemical Science Experimental Center of the China University of Science and Technology (Hefei). The ICP-MS instrument was an X Series 2 model. Sample pretreatment was undertaken in the Suzhou Graduate School (China University of Science and Technology). The procedure was as follows: first, 2 g of fresh coal sample was selected, ground and sieved (200-mesh) to obtain a powered sample; subsequently, the sample was digested with mixed acid (hydrofluoric acid, perchloric acid and nitric acid) on a hot plate and after cooling was made up to constant volume (25 mL). The sample digests were then sent to the Physical and Chemical Science Experimental Center (China University of Science and Technology) for ICP-MS analysis. Two coal certified reference materials (SARM20) were selected as standard samples for instrument calibration and to perform method development experiments and checks including blank tests. For determination of As and Se, measurements were performed using atomic fluorescence spectrometry (AFS) (model AFS-230a) analysis. The ICP-MS and AFS analyses were performed on the three coal samples previously mentioned.

4 Results and discussion

4.1 Basis of cleanliness grade evaluation

4.1.1 Coal quality characteristics

Based on a systematic assessment of the coal quality data for the No. 13-1 coal seam of the Huainan Coalfield and the supplementary coal quality data (Table 2), it was concluded that the A_d for the coal seam was in the range 19.56%–24.94% (average value 22.51%), hence the coal was considered to be a low-medium ash coal; in addition, the coal distribution was relatively high in the north and south, and low in the middle section (Fig. 2).

The $S_{t,\ d}$ values were in the range 0.26%–0.60% (average 0.37%), indicating that the coal was of the super low S coal grade, and having the characteristics of being relatively low in the middle and high in the other sections (Fig. 3). The coefficient of variation (CV) for the A_d and $S_{t,\ d}$ values of the No. 13-1 coal seam were less than 1 (Table 2). The preliminarily conclusion was that that the quality of the coal seam was not influenced by the local environment at the time of coal formation.

4.1.2 Contents of hazardous trace elements

As can be seen from Fig. 4, the contents of Cr, Mn and Ni elements are generally high, with average values being greater than 20 μg/g; in the case of Mn the average value reached 80 μg/g. Arsenic, Se, Hg and other trace elements were generally less than 10 μg/g; the element with the lowest concentration was Hg, the average value being 0.43 μg/g. To reflect the degree of enrichment for the trace elements in the coals, the ratio (*R*) of the arithmetic mean of the element content in the coal for the study area to the average value for the element in coal for the world average was calculated. An R value > 4 indicates a relatively high enrichment factor, whereas an R value < 0.2 indicates a relatively low enrichment factor; *R* values lying within the specified limits were considered to have a normal element content (Ren et al. 1999). The enrichment factors for the hazardous trace elements in coal of the No. 13-1 coal seam are shown in Fig. 5. As can be seen from Fig. 5, the enrichment factor data for the trace elements in the coals of the Huainan Coalfield are quite similar to those reported by Chen et al. (2011), i.e., the elements are considered to occur at normal element levels, except for Se and Hg which exhibit a high degree of enrichment.

The trace element contents of the coals from the No. 13-1 coal seam are presented in Table 2. It can be seen that the Cr contents ranged from 17.50 to 58.65 μg/g (average 38.75 μg/g; $n = 48$), the contents being higher than those in the study of Chen et al. (2011) (average 27.52 μg/g; $n = 30$); the contents of Mn ranged from 12.27–75.50 μg/g (average 43.73 μg/g; $n = 39$); the contents of Ni ranged from 12.03–85.15 μg/g (average 32.04 μg/g; $n = 48$), higher than Chen et al. (2011) (22.39 μg/g; $n = 30$); the contents of As ranged from 1.37 to 20.72 μg/g (average 6.20 μg/g; $n = 42$), lower than Chen et al. (2011) (9.20 μg/g; $n = 30$); the contents of Se ranged from 3.02–24.36 μg/g (average 8.8 μg/g; $n = 42$), higher than Chen et al. (2011) (3.44 μg/g; $n = 10$); the contents of Cd ranged from 0.05 to 6.21 μg/g (average 0.85 μg/g; $n = 44$); the contents of Hg ranged from 0.13 to 0.80 μg/g (average 0.34 μg/g; $n = 8$);

Table 2 Coal quality and hazardous trace elements in the No. 13-1 coal seam of the Huainan Coalfield

Mining area	Coal mine	A_d	$S_{t,d}$	Cr		Mn		Ni		As		Se		Cd		Hg		Pb	
		(%)	(%)	Content (μg/g)	Number	Content (μg/g)	Number	Content (μg/g)	Number	Content (μg/g)	Number	Content (μg/g)	Number	Content (μg/g)	Number	Content (μg/g)	Number	Content (μg/g)	Number
Huainan	XZZ	21.41	0.60	47.80	2	nd	0	85.15	2	3.74	2	8.51	2	0.07	2	nd	0	nd	0
Panxie	PY	21.91	0.30	40.00	6	58.00	6	22.80	6	2.20	6	7.80	6	0.18	6	0.35	6	19.40	6
	PE	21.94	0.50	48.00	5	69.60	5	27.00	5	2.64	5	9.36	5	0.22	5	nd	0	23.28	5
	PS	24.27	0.35	52.10	10	75.50	10	25.38	10	2.72	10	8.88	10	0.50	10	nd	0	23.68	10
	PJ	24.73	0.36	26.65	6	nd	0	12.03	6	nd	0	nd	0	0.05	6	nd	0	13.90	6
	ZJ	24.94	0.35	35.25	11	55.72	11	28.10	11	2.75	11	9.75	11	0.23	11	nd	0	18.43	11
	DJ	22.51	0.31	17.50	1	24.62	1	22.50	1	3.70	1	3.53	1	0.50	1	nd	0	13.50	1
	ZHJ	23.25	0.32	44.41	5	30.47	5	37.89	5	2.40	5	15.29	5	6.21	1	0.60	1	0.12	1
Fudong	LZ	19.56	0.26	31.56	1	28.92	1	28.56	1	20.72	1	3.02	1	0.28	1	nd	0	8.74	1
	KZD	20.6	0.34	38.60	1	20.29	1	25.18	1	18.76	1	6.53	1	0.22	1	0.13	1	15.93	1
Minimum		19.56	0.26	17.50		12.27		12.03		1.37		3.02		0.05		0.13		0.12	
Maximum		24.94	0.60	58.65		75.50		85.15		20.72		24.36		6.21		0.80		23.68	
Mean		22.51	0.37	38.75		43.73		32.04		6.20		8.80		0.85		0.43		15.22	
Total		nd	nd	48		39		48		42		42		44		8		42	
Standard deviation		1.69	0.10	12.16		22.84		20.31		7.19		5.94		1.89		0.34		7.41	
Coefficient of variation		0.08	0.26	0.31		0.52		0.63		1.16		0.68		2.23		0.80		0.49	

The above data include data collected from the literature and the measured data. Except the measured data, the rest are collected data(Huang et al. 2000; Chen et al. 2009; Chen 2013; Liu et al. 2009; Kong 2007; Wu 2006; Ping 2016; Yan 2014; Chen et al. 2011, 2014; Sun et al. 2010a, b; Zhou et al. 2014; Yang et al. 2012; Yan et al. 2014; Ding et al. 2018)

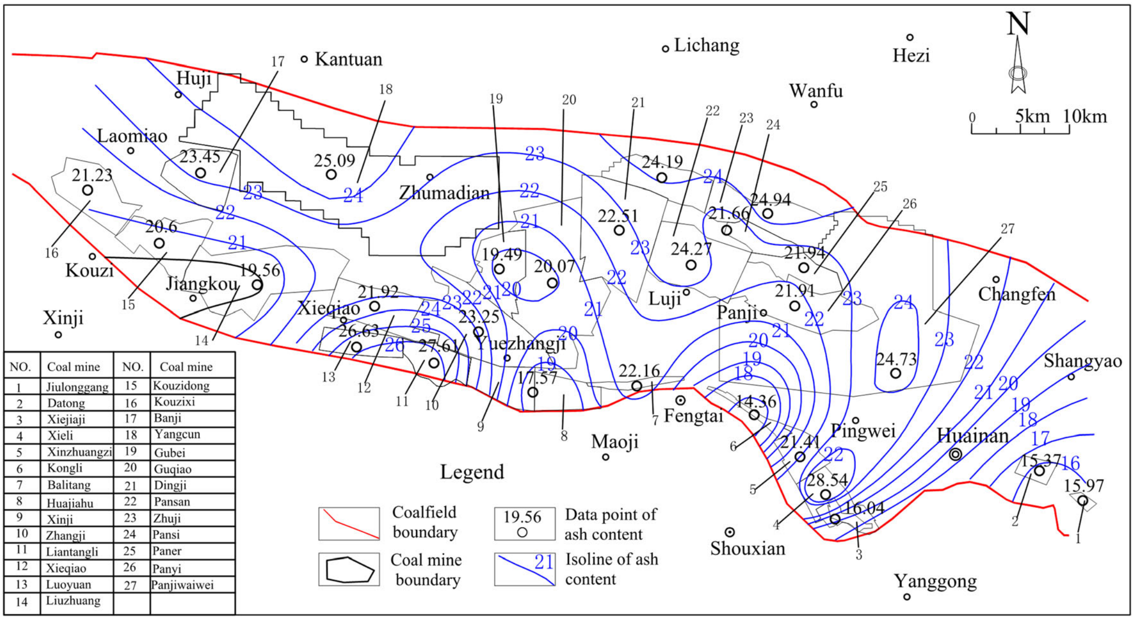


Fig. 2 Isoline map for coal ash content in the No. 13-1 coal seam of the Huainan Coalfield

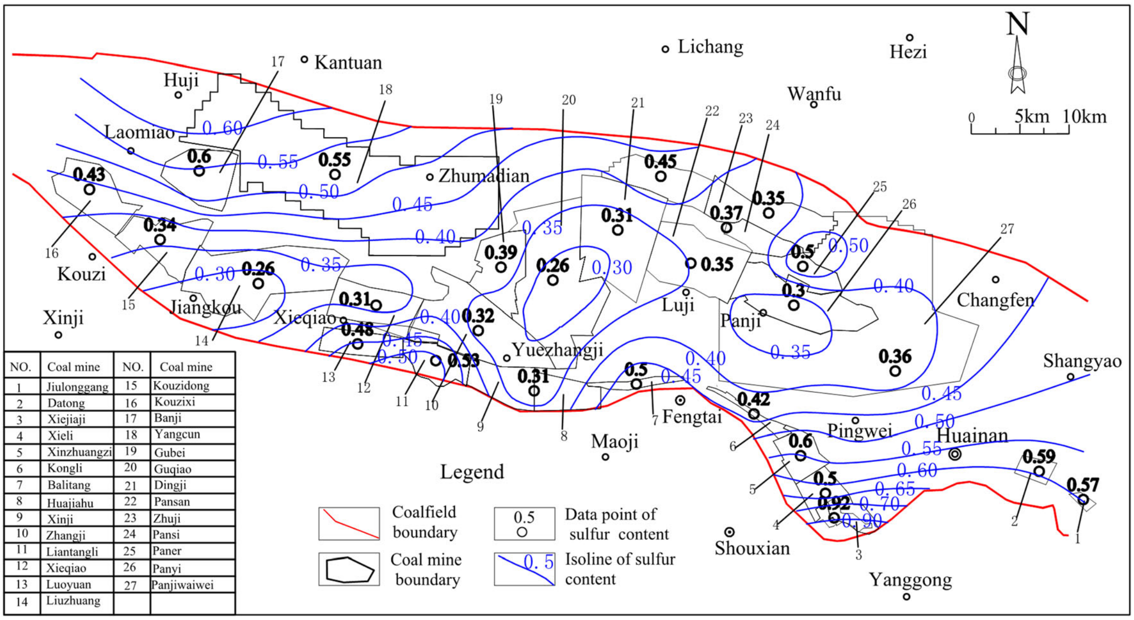


Fig. 3 Isoline map for the sulfur content of the No. 13-1 coal seam of the Huainan Coalfield

the contents of Pb ranged from 0.12–23.68 μg/g; average 15.22 μg/g; $n = 42$), higher than Chen et al. (2011) (12.89 μg/g; $n = 18$). It can be further seen from inspection of Table 2 that the CVs for all elements were less than 1, except for As and Cd. Therefore, it can be inferred that As and Cd in coals of the No. 13-1 coal seam were greatly affected by the past sedimentary environment of the Huainan Coalfield and the later superimposed geological processes.

4.2 Evaluation of cleanliness

4.2.1 Selection of evaluation factors

At present, the determination of the A_d and S limits in coal mainly refers to the division of the cleanliness grades in the GB/T15224–2010 standard. The determination of the limits for hazardous trace elements in coal is quite complicated, and the reference standards vary depending on the purposes of research. The details are as follows:

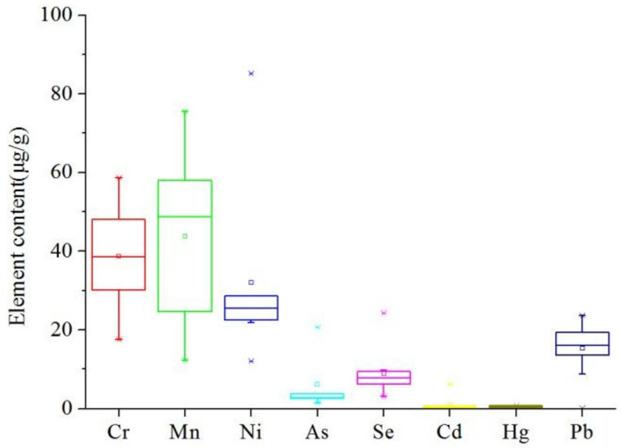


Fig. 4 Box diagram plots for hazardous trace elements in the No. 13-1 coal seam

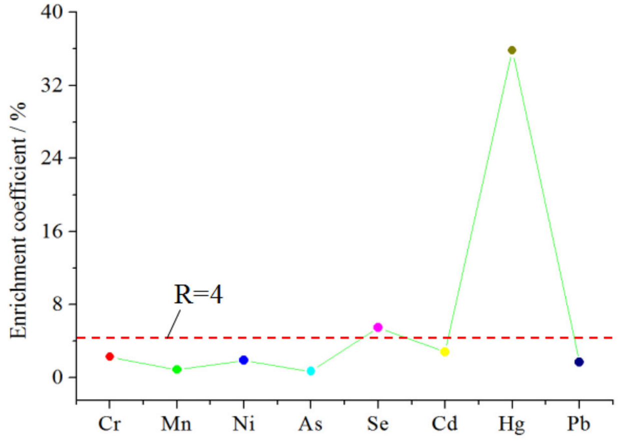


Fig. 5 Enrichment factor (R) values for hazardous trace elements in the No. 13-1 coal seam

(1) For atmospheric environmental protection standards, the Clean Air Supplement Act (1990) enacted by the US Congress specifies 11 hazardous trace elements in coal, namely Cr, Mn, Co, Ni, As, Be, Se, Cd, Sb, Hg and Pb. In China, the "Comprehensive Emission Standard for Air Pollutants" (GB16297–1996) stipulates the maximum allowable concentrations for five hazardous trace elements including Cr, Ni, Cd, Hg and Pb elements in coal. This air quality standard represents the highest concentrations of hazardous trace elements in the atmosphere for China.

(2) For environmental protection standards for waters, the U.S. Environmental Protection Agency (1976), China's "Sanitary Standard for Drinking Water" (GB5749–85), "the Highest Concentration of Hazardous Substances in Surface Water" (TJ35–79), the "Farmland Irrigation Water Quality Standards" (GB5089–1992) and the "Comprehensive Wastewater Discharge Standards" (GB8978–1996) all specify the corresponding maximum allowable concentrations of hazardous trace elements.

(3) For environmental protection standards for soil, China's "Soil Environmental Quality Standard" (GB15618–1995) stipulates the maximum allowable concentrations of eight hazardous trace elements, namely Cr, Ni, Cu, Zn, As, Cd, Hg and Pb.

Based on the current research status of hazardous trace elements in coals of the Huainan Coalfield and related environmental protection standards, A_d, $S_{t,d}$, Cr, Mn, Ni, As, Se, Cd, Hg and Pb were selected as evaluation factors for the grading of coal cleanliness. In view of the extremely low contents of F and Cl in coals of the Huainan Coalfield (F: 60.04–144.91 μg/g; Cl: 80–380 μg/g; these concentrations correspond to super-low to low fluorine coal and super-low chlorine coal, respectively), F and Cl were not used as evaluation indices in this study.

4.2.2 *Quantification of evaluation factors*

To quantitatively evaluate the coal cleanliness grade of coal resources, it is necessary to quantify the above evaluation factors and calculate the theoretical limits for the environmental concentrations for each evaluation factor. Based on the relevant environmental standards for each evaluation factor and the average value for each evaluation factor in the earth's crust, in world-average coal and in Chinese coal, the concentration limits for the above evaluation factors were calculated.

In this study, the quantification of A_d was based on the corresponding national standards; the quantification of $S_{t,d}$, Cr, Ni, Cd, Hg and Pb was based on the "Comprehensive Emission Standard for Air Pollutants" (GB 16297–1996); the quantification of Mn, As and Se was based on miscellaneous environmental standards, such as the "Concentration Standard of Hazardous Substances in the Air of Production Workshops." The volatilization rates for the hazardous trace elements were from the results of Tang et al. (2006) and Hu et al. (2018).

(1) Ash yield (A_d)
According to the national standard for A_d (GB/T 15224.1–2018), an A_d of 10% was used as the standard (allowable concentration value) to distinguish between super good clean coal and good clean coal. An A_d of 30% was taken as the standard (maximum concentration value) to distinguish between relatively poor clean coal and poor clean coal. The above division of the clean coal concentration limits was in line with the actual situation of the Huainan Coalfield.

(2) Sulfur content($S_{t,\ d}$)

According to the national standard for $S_{t,\ d}$ (GB/T 15224.2–2010), the $S_{t,\ d}$ for coal of the Huainan Coalfield coal corresponds mainly to super-low to low sulfur coal. Zhang (1999) reported that the toxicity limit for $S_{t,\ d}$ in coal was 1.5%, and the theoretical allowable concentration value was 0.57%. Therefore, in this study, 0.5% was used as the standard (allowable concentration value) to distinguish between super good clean coal and good clean coal, and 1.5% was used as the standard (maximum concentration limit) to distinguish between relatively poor clean coal and poor clean coal.

(3) Cr

According to Wedephol (1995), Swaine (1994), Ketris and Yudovich (2009), Ren et al. (2006) and Dai et al. (2011), the abundance of Cr in the earth's crust is 126 µg/g; the abundance of Cr in the world's coal is 17 µg/g, and the average content of Cr in Chinese coal is 15.4 µg/g. In addition, studies (Li et al. 1993; Liao 1989) have shown that the content of Cr in coal is hazardous if the concentration exceeds 100 µg/g. For comparison purposes, the concentration of Cr in soil is about 20–40 µg/g, which clearly would stimulate the growth of corn. Therefore, it was considered appropriate to set the allowable and the maximum concentrations of Cr in coal to be 6 and 40 mg/kg, respectively.

(4) Mn

According to Swaine (1994), Ketris and Yudovich (2009), Ren et al. (2006) and Dai et al. (2011), the abundance of Mn in world coal was 50 µg/g, while the average content of Mn in Chinese coal was 271.2 µg/g. With reference to the air quality standard in the former Czech Republic (1969), the concentration of Mn in the working environment should not exceed 2 mg/kg, and the allowable concentration of Mn in coal was calculated to be 91 mg/kg. In the case of the equivalent standard in the United States (1974), the highest concentration of Mn in the working environment should not exceed 5 mg/kg, and the allowable concentration of Mn in coal was calculated to be 228 mg/kg. Therefore, it was considered appropriate to set the allowable and maximum concentrations of Mn in coal to be 90 mg/kg and 230 mg/kg, respectively.

(5) Ni

According to Wedephol (1995), Swaine (1994), Ketris and Yudovich (2009), Ren et al. (2006) and Dai et al. (2011), the abundance of Ni in the earth's crust was 56 µg/g; the abundance of Ni in world coal was 17 µg/g, while the average content of Ni in Chinese coal was 13.7 µg/g. Referring to the allowable concentration value (4.3 mg/kg) of Ni in the atmospheric environment in "Comprehensive Emission Standards for Air Pollutants" (GB16297–1996), the allowable concentration of Ni in coal was calculated to be 100 mg/kg. According to China's "Soil Environmental Quality Standard" (GB15618–1995), the maximum allowable value for Ni in soil is 200 mg/kg. Therefore, it was deemed appropriate to set the allowable and maximum concentrations of Ni in coal as 100 and 200 mg/kg, respectively.

(6) As

According to Wedephol (1995), Ketris and Yudovich (2009) and Dai et al. (2011), the abundance of As in the earth's crust is 1.7 µg/g; the abundance of As in world coal is 9.0 µg/g, while the average content of As in Chinese coal is 3.79 µg/g. Zhang et al. (1999) reported that if the content of As in coal was more than 10 mg/kg, it would cause environmental pollution. The state stipulated that the content of As in coal for food industry-related use should not be greater than 8 mg/kg, but a slightly higher content of As in coal for non-food industry applications was acceptable. Therefore, the allowable concentration of As in coal was calculated to be 4.72 mg/kg. It was thus deemed appropriate to set the allowable and maximum concentrations of As in coal as 5 and 20 mg/kg, respectively.

(7) Se

According to Wedephol (1995), Ketris and Yudovich (2009) and Dai et al. (2011), the abundance of Se in the continental crust is 0.12 µg/g; the average content of Se in world coal is 1.6 µg/g, and the average content of Se in Chinese coal is 2.47 µg/g. Given that an environmental standard for Se has not been established yet, the allowable and maximum concentration for Se were set as 3 and 25 mg/kg, respectively, in accord with the research data of Yang et al. (2011).

(8) Cd

According to Wedephol (1995), Swaine (1994), Ketris and Yudovich (2009) and Zhao (1997), the abundance of Cd in the earth's crust is 0.10 µg/g; the abundance of Cd in world coal is 0.30 µg/g, while the average content of Cd in Chinese coal is 0.46 µg/g. Referring to the allowable concentration value (0.1 mg/kg) for Cd in the Comprehensive Emission Standards for Air Pollutants (GB16297–1996), the allowable concentration of Cd in coal was calculated to be 1.88 mg/kg. Chen and Lu (1989) reported that the minimum concentration of Cd in foodstuffs which was toxic was 13 mg/kg. However, considering the low content of Cd in Chinese coal and world coal, it was considered appropriate to set the allowable and maximum concentrations of Cd in coal as 1.0 and 5.0 mg/kg, respectively.

(9) Hg

According to Wedephol (1995), Ketris et al. (2009) and Dai et al. (2011), the abundance of Hg in the continental crust is 0.04 μg/g, the average content of Hg in world coal is 0.012 μg/g, and the average content of Hg in Chinese coal is 1.37 μg/g. The allowable concentration of Hg in the atmospheric environment is 0.012 mg/m^3 as stipulated in the "Comprehensive Emission Standard for Air Pollutants" (GB16297–1996), and the calculated allowable concentration of Hg in coal is 0.127 mg/kg. Further, Zhang et al. (1999) considered that the toxicity limit of Hg in coal was 0.5 mg/kg. Thus, it was deemed appropriate to set the allowable and maximum concentrations for Hg in coal as 0.13 and 0.50 mg/kg, respectively.

(10) Pb

According to Wedephol (1995), Swaine (1994), Ketris and Yudovich (2009), Ren et al. (2006) and Dai et al. (2011), the abundance of Pb in the earth's crust is 14.8 μg/g; the abundance of Pb in world coal is 9.0 μg/g, and the average content of Pb in Chinese coal is 15.1 μg/g. The allowable concentration of Pb in the atmospheric environment is 0.7 mg/m^3 as stipulated in China's "Comprehensive Emission Standard for Air Pollutants" (GB16297–1996). The allowable concentration of Pb in coal was calculated as 13.17 mg/kg. In addition, according to Li et al. (1993) and Liao et al. (1989), the content of Pb in common plants is about 40 mg/kg, therefore, the rice yield would be reduced if the water used for irrigation was contaminated with Pb at a concentration of 50 mg/L. Therefore, the allowable and maximum concentrations of Pb in coal were deemed to be 13 and 50 mg/kg, respectively.

The quantitative criteria for the environmental standards are specified in Table 3.

4.2.3 *Establishment of evaluation model for the coal cleanliness grade*

Many different classification schemes have been issued for the grading of coal resources, including: (1) A five category classification scheme (China General Administration of Coal Geology); (2) a four-categories classification scheme (Tang et al. 2006); and a six-categories classification scheme (Yang et al. 2011). In this study, the cleanliness grading of coal resources is divided into six grades, that is, super clean coal, good clean coal, relatively good clean coal, medium clean coal, relatively poor clean coal, and poor clean coal.

There are many methods in use to evaluate the cleanliness grade of coal resources, such as a fuzzy comprehensive evaluation method, a neural network method, an index method, a grey clustering method and a generalized contrast weighted scale index method (GCWSIM) (Li 2000; Tang et al. 2006). Among them, the fuzzy comprehensive evaluation method and the GCWSIM are widely used. In this study, the GCWSIM (Li 2000) was used as a method to evaluate the cleanliness grade of coal resources. This index method, as part of the evaluation process, takes account of the impact on the environment of the degree of air

Table 3 Environmental reference standards and calculated results for the evaluation factors (after Tang et al. 2006 and Hu et al. 2018)

Factor	Allowable concentration (mg/m^3)	National standard	Volatilization rate (%)	Theoretical concentration limit (mg/kg)	Allowable concentration actually used (mg/kg)	The highest concentration actually used (mg/kg)
A_d	–	–	–	–	10	30
$S_{t,d}$	–	–	–	–	0.5	1.5
Cr	0.07	Air	10	6.1	6	40
Mn	2.0	Workshop	20	91.3	90	230
Ni	4.3	Air	40	100.6	100	200
As	0.3	Workshop	60	4.72	5	10
Se	0.1	–	70	1.35	1.5	5
Cd	0.1	Air	50	1.88	1.0	5
Hg	0.012	Air	90	0.127	0.13	0.5
Pb	0.70	Air	50	13.17	13	50

Table 4 Concentration limits (mg/kg) for six levels of cleanliness(C_j) and the comprehensive index values I_j for the evaluation factors

Factor	Background value/C_{jo}	Level I		Level II		Level III		Level IV		Level V		Level VI
		C_{j1}	I_{j1}	C_{j2}	I_{j2}	C_{j3}	I_{j3}	C_{j4}	I_{j4}	C_{j5}	I_{j5}	C_{j6}
A_d (%)	5	10	0.301	15	0.477	25	0.602	30	0.778	40	0.903	50
S (%)	0.3	0.5	0.269	0.7	0.447	1.0	0.635	1.2	0.731	1.5	0.848	2.0
Cr	4	6	0.146	10	0.330	15	0.477	25	0.661	40	0.830	64
Mn	70	90	0.177	115	0.349	145	0.512	180	0.664	230	0.837	290
Ni	80	100	0.203	120	0.369	140	0.509	170	0.686	200	0.834	240
As	3.5	5	0.111	7	0.278	10	0.455	15	0.656	20	0.799	30
Se	2	3	0.132	5	0.299	9	0.490	15	0.657	25	0.823	43
Cd	0.7	1.0	0.150	1.5	0.321	2.2	0.483	3.3	0.654	5	0.829	7.5
Hg	0.09	0.13	0.179	0.18	0.338	0.26	0.517	0.36	0.676	0.50	0.836	0.70
Pb	8	13	0.224	18	0.374	25	0.525	36	0.693	50	0.845	70
I			0.190		0.358		0.544		0.696		0.847	

Table 5 Comprehensive indices I for classification of coal resource cleanliness

Cleanliness grade		Comprehensive index I
I	Super clean coal	$I \in [-\infty, 0.190]$
II	Good clean coal	$I \in [0.190, 0.358]$
III	Relatively good clean coal	$I \in [0.358, 0.544]$
IV	Medium clean coal	$I \in [0.544, 0.696]$
V	Relatively poor clean coal	$I \in [0.696, 0.847]$
VI	Poor clean coal	$I \in [0.847, +\infty]$

Table 6 Evaluation results for the cleanliness grade of the No. 13-1 coal seam of the Huainan Coalfield

Mining area	Coal mine	Comprehensive index I	Cleanliness grade
Huainan	XZZ	0.600	IV
Panxie	PY	0.624	IV
	PE	0.617	IV
	PS	0.618	IV
	ZJ	0.530	III
	ZHJ	0.584	IV
	DJ	0.373	III
	PJ	0.813	V
Fudong	LZ	0.785	V
	KZD	0.607	IV

pollution in the process of coal combustion. The degree of impact is expressed by an index, which ranges from the background concentration C_{jo} of air pollutants to the maximum concentration limit C_{jd}. The change of pollutants (equal ratio basis) corresponds to the change of the sub-index (equal ratio basis), which is the sub-index scale (generally expressed by K_j), expressed specifically as:

$$K_j = \frac{\lg(C_{jk} / C_{jo})}{lga_j} \tag{1}$$

where, A is the measured concentration of element j; B is the background concentration of element j; C is the importance ratio of the adjacent sub-index level of elements; C_{jk} is the measured concentration value of element j; C_{jo} is the background concentration of element j; and a_j is the importance ratio of the adjacent sub-index level of elements,

$$a_j = C_{jk} / C_{jo}{}^{1/9} \tag{2}$$

The scaling exponent of Eq. (2) is normalized to obtain:

$$I_j = \frac{1}{9} \quad K_j = \frac{\lg(C_{jk}) - \lg(C_{jo})}{\lg(C_{jd}) - \lg(C_{jo})} \tag{3}$$

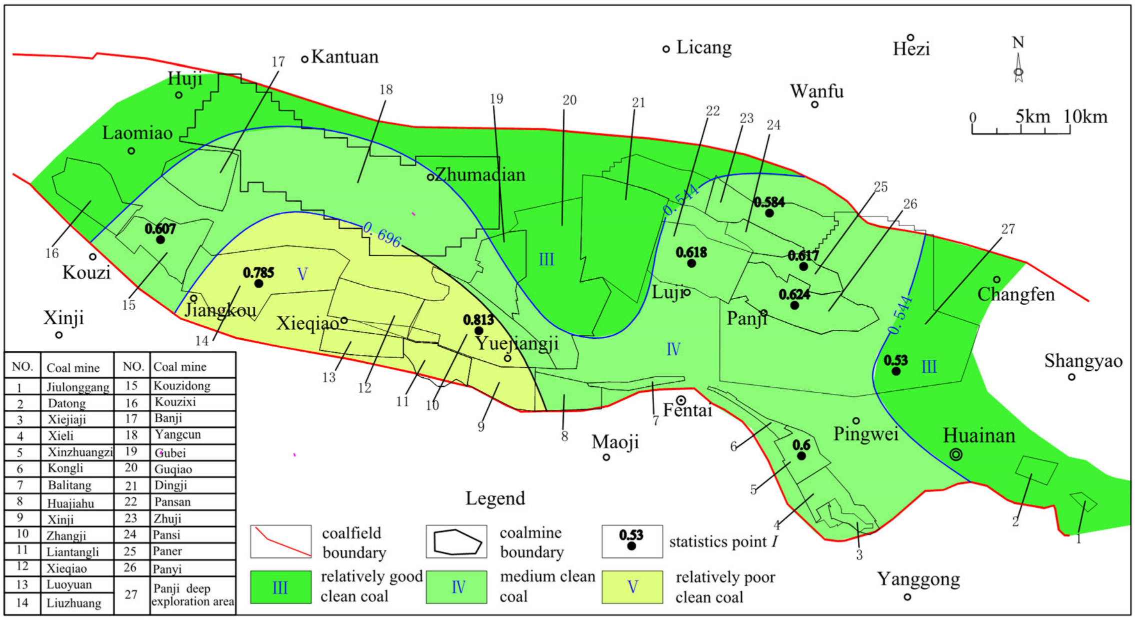
The weight W_j of each index factor was obtained by the following formula where if $I_j < 0$, W_j takes the value of 0:

$$W_j = \begin{cases} 2^{p-1} I_j^p & 0 \leqslant I_j \leqslant 0.5 \\ 1 - 2^{p-1} 1 - I_j^p & 0.5 < I_j \leqslant 1 \\ 1 + 2^{p-1} I_j - 1^p & I_j > 1 \end{cases} \tag{4}$$

where, p is an adjustable parameter and is generally taken as 1/2. W_j is normalized to get $W_j{}^*$, and the composite index I is obtained as:

$$I = \sum_{j=1}^{m} W_j{}^* I_j \tag{5}$$

According to the allowable and the highest concentration limit values for each evaluation factor, the scaling index method was adopted to divide approximately the



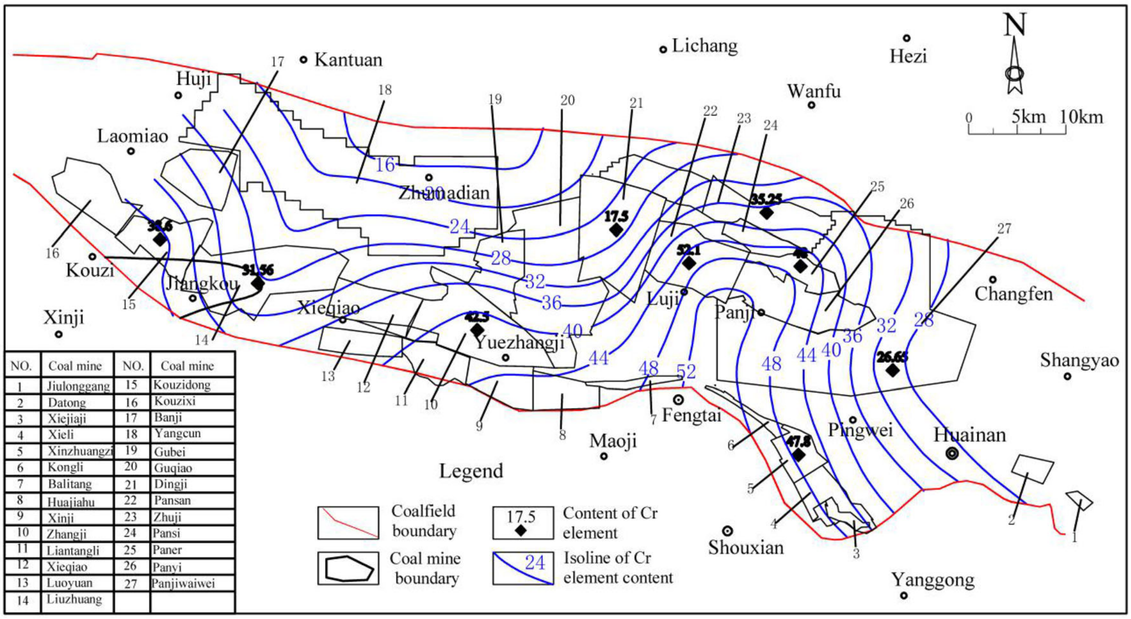
NO.	Coal mine	NO.	Coal mine
1	Jiulonggang	15	Kouzidong
2	Datong	16	Kouzixi
3	Xiejiaji	17	Banji
4	Xieli	18	Yangcun
5	Xinzhuangzi	19	Gubei
6	Kongli	20	Guqiao
7	Balitang	21	Dingji
8	Huajiahu	22	Pansan
9	Xinji	23	Zhuji
10	Zhangji	24	Pansi
11	Liantangli	25	Paner
12	Xieqiao	26	Panyi
13	Luoyuan	27	Panji deep exploration area
14	Liuzhuang		

Fig. 6 Regional distribution map of the cleanliness grade for the No. 13-1 coal seam of the Huainan Coalfield

concentration limit values at all levels into equal ratios, and the six-level concentration limit values for each evaluation factor were obtained (Table 4). The comprehensive index *I* can be calculated by Eqs. (1), (2) and (3) as given in Table 5.

4.2.4 Model application

Table 6 shows the evaluation results for the cleanliness grade for the No. 13-1 coal seam of the Huainan Coalfield. It can be seen that the coal resources of the coalfield generally belong to the relatively good-medium clean coal grade (grade III–IV), while some areas have relatively poor clean coal (grade V). Among them, the relatively good clean coal is mainly distributed in the Dingji-Guqiao area in the central part of the coalfield and the eastern area around the Panji coalmine; medium clean coal is mainly distributed in the Panji, Xiejiaji, Xinji and Kouzidong coalmine areas; relatively poor clean coal is mainly



NO.	Coal mine	NO.	Coal mine
1	Jiulonggang	15	Kouzidong
2	Datong	16	Kouzixi
3	Xiejiaji	17	Banji
4	Xieli	18	Yangcun
5	Xinzhuangzi	19	Gubei
6	Kongli	20	Guqiao
7	Balitang	21	Dingji
8	Huajiahu	22	Pansan
9	Xinji	23	Zhuji
10	Zhangji	24	Pansi
11	Liantangli	25	Paner
12	Xieqiao	26	Panyi
13	Luoyuan	27	Panjiwaiwei
14	Liuzhuang		

Fig. 7 Regional distribution map of the Cr content for the No. 13-1 coal seam of the Huainan Coalfield

distributed in the Luoyuan–Liantangli–Xieqiao–Liu Zhuang areas (Fig. 6).

Furthermore in the areas of the coalfield corresponding to the medium clean coal and relatively poor clean coal grades, the content of Cr exceeds the maximum concentration limit in most coal seams (Fig. 7). As revealed in Fig. 7, the distribution of Cr is relatively high in the south and low in the north, which is consistent with the results of Fig. 6. In general, the contents of Se, As, Hg and Pb are close to or exceed the maximum concentration limits in some coal seams. Therefore, it is necessary to monitor closely these elements and assess the extent of air pollution due to these elements in combustion processes.

To sum up, although there are medium or relatively poor clean coal grades in the Huainan Coalfield, the most hazardous trace elements in these coal seams are mainly contained in the A_d of the coal. It is possible to attain an improved cleanliness grade after coal washing. In addition, considering that comprehensive utilization of coal resources in the Huainan Coalfield used to include production of traditional coking coal, oil refining coal, power coal and civil coal, etc., such uses are no longer considered to equate to a clean and efficient utilization of the coal resources. To realize clean and efficient utilization of coal resources in the Huainan Coalfield, the following needs to be addressed: first, technical innovations for comprehensive utilization of coal should be strengthened to improve coal combustion efficiency (such as washing technology, power plant desulfurization and dust removal technology); second, environmental monitoring of hazardous trace elements (especially Cr, Se, As, Hg and Pb) associated with coal combustion should be strengthened as a means to control and reduce air pollution; third, it is necessary to strengthen the processing technology for extraction of coal at depth, and introduce measures to convert in situ coal to clean energy (e.g., coal to gas, coal to oil), to realize clean and efficient utilization of coal resources in the true sense.

5 Conclusions

The main findings and conclusions of research are as follows:

(1) The contents of Cr, Mn, and Ni in the No. 13-1 coal seam of the Huainan Coalfield are relatively high with the average values being greater than 20 μg/g. The contents of Se and Hg are considered to be at a relatively high level, while the remaining trace elements are judged to be at a normal level.

(2) By using the GCWSIM and considering the element volatilization rate, an improved model for evaluating the cleanliness grade of coal resources in the Huainan Coalfield has been developed.

(3) The cleanliness grade of coal resources in the Huainan Coalfield was mainly grade III–IV, which corresponds to a relatively good-medium clean coal; grade V coal cleanliness (relatively poor clean coal) occurred in some areas. Overall, the evaluation results for coal resource cleanliness reflects the actual situation for the No. 13-1 coal seam of the Huainan Coalfield.

(4) The contents of Cr, As and Hg exceeded the maximum concentration limits for the relatively poor clean coal areas (Luoyuan–Liantangli–Xieqiao–Liu Zhuang area), therefore, it is necessary to monitor closely air pollution associated with the release of the above elements in coal combustion and if necessary take corrective actions.

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