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Review

Distribution, Enrichment and Modes of Occurrence of Arsenic in Chinese Coals

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Abstract: Arsenic is one of the toxic trace elements in coals, which is harmful to both the ecological environment and human health. Based on published literature and the data obtained by our research group, a total of 5314 As concentrations of Chinese coals were analyzed. The arithmetic mean of arsenic content in Chinese coals is 6.97 mg/kg. Choosing the percentage of provincial coal resources in national coal resources as the weighting factor, the weighted average of arsenic content in Chinese coals is 5.33 mg/kg. The content of arsenic in Chinese coals increases from the north to the south. High arsenic content in coal primarily occurs in southwestern Yunnan and certain coalfields in the Guizhou Province. Additionally, arsenic is enriched in the coals from some regions, i.e., the western Yunnan, Guangxi, Tibet, southwestern Liaoning, Jilin, and Henan. The arsenic content in coals of different coal-forming periods shows an overall regularity: Paleogene and Neogene > Late Triassic > Late Permian > Late Jurassic and Early Cretaceous > Early and Middle Jurassic > Late Carboniferous and Early Permian. The modes of occurrence of arsenic in coals include sulfide-association, organic-association, arsenate-association, silicate-association, and soluble- and exchangeable-association. Generally, arsenic in Chinese coals exists predominantly in arsenic-bearing pyrite. Meanwhile, the organic arsenic content is relatively high in coal samples with a lower (<5.5 mg/kg) arsenic content and a low or medium ash yield (<30%).

Keywords: arsenic; Chinese coals; distribution; enrichment; modes of occurrence

1. Introduction

In 2016, China's coal production was 3.41 billion tons, which was 7.9% lower than 2015, accounting for 46.1% of the total world coal production. Additionally, the coal consumption was 3.82 billion tons, 50.6% of the world consumption [1,2]. The share of coal as China's primary energy source reached 62.0% in 2016 [2], and coal will locate in a dominant position of Chinese energy structure in the foreseeable future.

The toxic trace elements in coals can migrate into the atmosphere, hydrosphere and soil, causing environmental pollution and even harmful impacts on human health during coal utilization [3]. As one of the most toxic trace elements in coals, arsenic could be released into the atmosphere during coal combustion and preferentially enriched in fly ash [4]. The arsenic-rich fly ash could remain in the air for a long time and enter the human body through the respiratory system [5]. Finally, the fly ash precipitates on the earth's surface, resulting water and soil pollution. The amount of arsenic entering the soil cycle in the form of fly ash reached 2200 tons every year [6]. Global arsenic emission from coal combustion was 6240 tons per year [7]. The atmospheric arsenic emissions from coal combustion reached 1564 tons in China in 2005 [8]. Arsenic can also be enriched in coal-hosted rare-metal deposits and their corresponding coal combustion products, which could have adverse effects for both environment and human health [9–12].

The earliest study on arsenic in world coals could be traced back to the 1934 conducted by Goldschmidt and Peters [13]. Before determined by inductively coupled plasma mass spectrometry (ICP-MS), coal samples should be digested. The microwave digestion program, related to coal and coal-related materials, was outlined by Dai et al. [14]. And it was recently determined by ICP-MS using collision cell technology (CCT) in order to avoid disturbance of polyatomic ions [15,16]. Valkovic [17], Yudovich et al. [18], and Ketris and Yudovich [19] estimated the average content of arsenic in coal. The modes of occurrence of arsenic in coals, migration and transformation mechanisms were discussed [13,20,21]. Finkelman [22] assigned the confidence level for arsenic occurrence in coal in pyrite as 8. The methods for the modes of occurrence of trace elements in coals can be divided into direct and indirect methods, i.e., microscopic and spectral analysis, float and sink analysis, sequential chemical extraction, low temperature ashing + X-ray diffraction, and statistical analysis [23]. The Mega-pixel Synchrotron X-ray Fluorescence (MSXRF), X-ray Absorption Near Edge Structure (XANES), and Extended X-ray Absorption Fine Structure (EXAFS) have recently contributed to the modes of occurrence of arsenic in coals [24]. The sulfides-associated As dominate at high As content, while As_{org} dominates at a low As level [25]. Regarding pyrite-hosted arsenic in coal, it occurs as a solid solution [26].

In 1964, villagers in the Zhijin County of Guizhou Province suffered from chronically arsenism due to the high arsenic level in coals, attracting much attention on the arsenic in Chinese coals [27–32]. Besides Guizhou, the arsenism also occurred in the Yunnan and Shaanxi Provinces [33–36]. However, not all the Zhijin coals are rich in arsenic [31,37,38]. The mechanism of arsenic enrichment in Chinese coals was fully discussed by Zhao [30], Ren et al. [39], and Dai et al. [40]. There are five types, i.e., source-rock-controlled, marine-environment-controlled, hydrothermal-fluid-controlled, groundwater-controlled, and volcanic-ash-controlled [40].

Due to the complex geologic conditions, multiple coal-forming periods, and substantial epigenetic changes of Chinese coals, a comprehensive review on arsenic in Chinese coals is requisite. Thus, the average arsenic concentration of Chinese coals, spatial distribution, and modes of occurrence will be reviewed. In this paper, based on the published literature and the data of our research group, the arsenic in Chinese coals are completely reviewed, including arithmetic and weighted average of arsenic contents, distribution, abnormal enrichment, and modes of occurrence.

2. Content of Arsenic in Coals

2.1. Content of Arsenic in World Coals

In some countries, such as the United State of America, former Soviet Union, Australia, and Czech Republic, the geochemistry of arsenic in coal has completely investigated. The content of arsenic in American coal was 24.0 mg/kg (7676 samples), with a maximum of 2200 mg/kg [41]. The content of arsenic in the former Soviet Union coals was 25 mg/kg [42]. It reached 32.7 mg/kg in the Donetz coalfield [43]. Swaine [13,44] calculated that the arsenic contents in Australian coals were 1.50 and 2.00 mg/kg, respectively. Based on 9172 samples in the Bohemia basin, the arsenic content of coals from the northern Czech Republic ranged from 0.10 to 757 mg/kg, with an arithmetic mean of 39.9 mg/kg [45]. Furthermore, Pesek et al. [46] calculated that the content of arsenic in Czech coal increased from <0.10 to 2020 mg/kg, with an average of 209 mg/kg (23,601 coal samples). Yudovich and Ketris [25] estimated the average arsenic content of world coals and gave arsenic abundances in brown and bituminous coals of 7.6 ± 1.3 mg/kg and 9.0 ± 0.7 mg/kg, respectively. According to the panel on the trace element geochemistry of coal resource development related to health (PECH) statistics [47], the average content of arsenic in world coals was 5 mg/kg.

During the last two decades, several As-enriched coal deposits were reported from Turkey. The As enrichments in Turkish coal deposits was mostly related to the synchronous volcanic activity and leached surface waters [48–51]. The arsenic accumulation caused by synchronous volcanic activity was called "Turkish (volcanogenic) type" [25]. Furthermore, in certain places, epigenetic hydrothermal mineralization-related influence also elevated As-concentrations (up to 3854 mg/kg in Gediz coal) [52], and the multistage As-enrichments (up to 984 mg/kg) were reported [53].

2.2. Content of Arsenic in Chinese Coals

Chen et al. [27], analyzed 107 samples and reported the content of arsenic in Chinese coals, ranging from 0.32 to 97.8 mg/kg. Sun and Jervis [54] used 15 coal samples to calculate an arsenic content (0.06 to 124 mg/kg). Dou et al. [55] analyzed 732 samples in the Shenfu-Dongsheng mining area, concluding that the arsenic content in coal was in the range of 0.04–78.0 mg/kg, with an arithmetic mean of 1.77 mg/kg. Ren et al. [56] gave an arsenic content of 132 coal samples with 0.21–32,000 mg/kg (arithmetic mean of 276.6 mg/kg), which was ascribed to the introduction of some high arsenic coal samples from the southwestern China. There are significant variations of arsenic content in Chinese coals, especially some southwestern coal mines with elevated arsenic. For example, Ding et al. [32] reported that the arsenic in the Anlong coal from Guizhou Province reached 35,000 mg/kg, the greatest value around the world. Fortunately, the reserve of such extremely high-As coals is extraordinarily small. In view of its severe negative impact on environment, the local government has shut down these coal mines. It was not representative of mineable coalfields in China. Ren et al. [39] introduced a "reserves weight" method to recount 3453 Chinese coal samples and gave a new arithmetic mean of 3.80 mg/kg. Chen et al. [57] excluded the abnormally high arsenic coal samples in the calculation of common arsenic content in Chinese coals, ranging from 0.80 to 20.0 mg/kg (arithmetic mean of 4.00 mg/kg). Dai et al. [40] updated the value by analyzing 3386 coal samples, with an average of 3.79 mg/kg.

The high-arsenic coal occurs in southwestern Guizhou Province, resulting in a serious endemic arsenism caused by the domestic arsenic-rich coal combustion. More than 3000 patients in the mountainous region of southwestern Guizhou suffered from arsenosis [58]. The highest values given by Ding et al. (35,000 mg/kg) [32] and Zhao et al. (32,000 mg/kg) [59] were from southwestern Guizhou, so the general impression to many people is that all the coals in Guizhou are characterized by high arsenic. In fact, abnormal high-arsenic coal samples (such as 35,000 and 32,000 mg/kg) were from a small coal mine nearby, not a workable mine. The As-rich coals are not ubiquitous in the southwestern Guizhou [40], and the As-rich coals locate in a restricted area [60,61]. On the other hand, preventive measures for endemic arsenosis in Guizhou have achieved desired results [40].

In this paper, data from the published literature and our research group, with a total samples of 5314 from 30 provinces in China, are statistically analyzed. The arithmetic mean arsenic content of Chinese coals is 6.97 mg/kg, including many coal samples with a high arsenic content from the southwestern China, e.g., Lincang coals (117 mg/kg) in Yunnan Province [62] and Zhenfeng coals (54.8 mg/kg) in Guizhou Province [63]. The arsenic concentrations in Chinese coals are presented in Table 1.

The geographic distribution of arsenic in Chinese coal is extremely uneven, inducing a simple calculation of arithmetic mean cannot represent the common arsenic content of Chinese coals. Selecting the percentages of each province's coal resources in the Chinese total coal resources as weighting factors, the weighted averaging arsenic content in coals is calculated. This method can eliminate the difference between samples and geologic conditions caused by uneven sampling. Based on the predicted coal resources reported by the China Coal Geology Bureau [64], the resources weighted average arsenic content is 5.33 mg/kg, slightly higher than the data given by Dai et al. [40] and Ren et al. [39]. The results are listed in Tables 2 and 3, respectively. The arithmetic arsenic content in Chinese coals is far lower than that of American and Czech coals, but higher than that of Australian coals. However, with respect to the weighted arsenic content of Chinese coals, there is a little difference between China and world coals of 5 mg/kg [47].

Table 1. Arsenic concentration in Chinese coals (mg/kg). Data are quoted from references of [30,31,37–40,58,62,63,65–144].

Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source	Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source
Huainan, Anhui	3.57	22	C–P	Ge [65]	Daqingshan, Inner Mongolia	0.97	9	C–P	Dai et al. [113]
Huaibei, Anhui	0.6	7	C–P	Tang et al. [66]	Jungar Haerwusu, Inner Mongolia	0.6	29	C–P	Dai et al. [114]
Huaibei, Anhui	2.9	5	C–P	Tang et al. [66]	Jungar Chuancaogedan 5, Inner Mongolia	0.28	15	C–P	Yang et al. [115]
Huainan Xinzhuangzi, Anhui	10.7	2	C–P	Tang et al. [66]	Jungar Chuancaogedan 6, Inner Mongolia	0.56	7	C–P	Dai et al. [116]
Huainan Xinzhuangzi, Anhui	1.8	9	C–P	Tang et al. [66]	Jungar Heidaigou, Inner Mongolia	0.99	30	P_2	Dai et al. [117]
Huainan Liyi, Anhui	11.5	5	C–P	Tang et al. [66]	Yimin, Inner Mongolia	3	1	J_3–K_1	Ren et al. [39]
Huainan Panyi, Anhui	0.4	6	C–P	Tang et al. [66]	Dayan, Inner Mongolia	1	2	J_3–K_1	Ren et al. [39]
Huaibei Mengzhuang, Anhui	3.66	15	C–P	Zhao and Wu [67]	Huolinhe, Inner Mongolia	12.88	3	J_3–K_1	Bai [118]
Huainan, Anhui	2.02	17	C–P	Chen et al. [68]	Jalainur, Inner Mongolia	2.59	3	J_3–K_1	Ren et al. [39]
Huainan, Anhui	4.21	24	C–P	Tong et al. [69]	Yuanbaoshan, Inner Mongolia	2.95	2	J_3–K_1	Ren et al. [39]
Huainan Zhangji, Anhui	9.07	144	C–P	Li et al. [70]	Xidayao, Inner Mongolia	3.4	5	J_{1-2}	Ren et al. [39]
Huaibei, Anhui	1.51	12	C–P	Chen et al. [71]	Ordos Basin, Inner Mongolia	16.3	138	J_{1-2}	Li et al. [119]
Huainan, Anhui	1.58	24	C–P	Chen et al. [71]	Shengli, Inner Mongolia	8.46	30	J_3–K_1	Dai et al. [120]
Huainan, Anhui	4.43	22	C–P	Chen and Tang [72]	Shenfu-Dongsheng	1.77	732	J_{1-2}	Tang et al. [66]
Anqing and Tongling, Anhui	10.16	36	P_2	Qian and Yang [73]	Shenfu-Dongsheng	0.42	5	J_{1-2}	Tang et al. [66]
Beijing	1.62	3	C–P	Ren et al. [39]	Jiangoushan, Ningxia	0.55	1	C–P	Ren et al. [39]
Daanshan, Beijing	1	1	J_{1-2}	Ren et al. [39]	Shizuishan, Ningxia	1.57	8	C–P	Dai [102]
Changhe, Chongqing	10.02	5	P_3	Wang [74]	Shitanjing, Ningxia	0.97	11	C–P	Dai [102]
Songzao, Chongqing	3.17	32	P_2	Ren et al. [39]	Tongxin, Ningxia	1	3	C–P	Dai [102]
Nantong, Chongqing	3.08	21	P_2	Ren et al. [39]	Rujigou, Ningxia	0.74	5	J_{1-2}	Song [121]
Zhongliangshan, Chongqing	4.12	2	P_2	Ren et al. [39]	Ciyaobao, Ningxia	10.62	2	J_{1-2}	Li et al. [119]
Tianfu, Chongqing	4.4	3	P_2	Ren et al. [39]	Yuka, Qinghai	1.34	1	J_{1-2}	Ren et al. [39]
Chuandong, Chongqing	5.24	5	P_2	Ren et al. [39]	Mole, Qinghai	2.77	1	J_{1-2}	Ren et al. [39]
Chuandongnan, Chongqing	7.4	2	P_2	Ren et al. [39]	Jiangcang, Qinghai	3.05	1	J_{1-2}	Ren et al. [39]
Chongqing	3.07	1	P_2	Luo et al. [75]	Datong, Qinghai	3.62	1	J_{1-2}	Ren et al. [39]
Donglin and Nantong, Chongqing	2.547	32	P_2	this study	Tibet Plateau	0.788	16	J_{1-2}	Dai et al. [122]
Songzao, Chongqing	9.14	4	P_2	Dai et al. [76]	Huangxian, Shandong	2.9	2	E–N	Ren et al. [39]
Songzao, Chongqing	8.06	4	P_2	Dai et al. [76]	Feicheng and Xinwen, Shandong	11.4	17	C–P	Zen et al. [123]
Songzao, Chongqing	5.84	5	P_2	Dai et al. [76]	Feicheng and Xinwen, Shandong	1.57	6	C–P	Zen et al. [123]
Songzao, Chongqing	9.11	4	P_2	Dai et al. [76]	Zibo, Shandong	7.9	1	C–P	Tang et al. [66]
Songzao, Chongqing	5.42	4	P_2	Dai et al. [76]	Chaili, Shandong	3.5	1	C–P	Tang et al. [66]
Songzao, Chongqing	9.52	4	P_2	Dai et al. [76]	Taozhuang, Shandong	0.45	2	C–P	Tang et al. [66]
Songzao, Chongqing	25.7	5	P_2	Dai et al. [76]	Zaozhuang, Shandong	5.6	10	C–P	Tang et al. [66]
Chuandong, Chongqing	9.2	10	P_3	Ren et al. [39]	Jibei, Shandong	19.62	56	C–P	Hu [124]
Yongrong, Chongqing	10.43	17	P_3	Ren et al. [39]	Zaozhuang, Shandong	7.28	21	C–P	Chen et al. [71]
Changhebian, Chongqing	10.03	5	P_3	Wang et al. [77]	Jining, Shandong	2.34	38	C–P	Liu [125]
Moxinpo, Chongqing	2.27	4	P_2	Dai et al. [78]	Juye, Shandong	2.25	49	C–P	Zhao et al. [89]
Moxinpo, Chongqing	10.7	4	P_2	Dai et al. [78]	Xinwen, Shandong	1.67	7	C–P	Zhang [126]
Yongan, Fujian	12	3	P_2	Lu et al. [79]	Feicheng, Shandong	2.3	5	C–P	Zen et al. [123]
Fujian	7.5	2		Cui and Chen [80]	Tengxian, Shandong	0.8	1	C–P	Ren et al. [39]
Zhangye, Gansu	1.38	4	J_{1-2}	Ren et al. [39]	Taozao, Shandong	3.45	13	C–P	Chen and Tang [72]
Ankou, Gansu	1.5	2	J_{1-2}	Ren et al. [39]	Yanzhou, Shandong	1.79	6	C–P	Liu et al. [127]
Dayou, Gansu	6.43	2	J_{1-2}	Ren et al. [39]	Liaocheng, Shandong	7.9	1	C–P	Ren et al. [39]

Table 1. *Cont.*

Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source	Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source
Huating, Gansu	3.33	2	J_{1-2}	Ren et al. [39]	Zibo and Taozhuang, Shandong	4.12	14	C–P	Chen et al. [57]
Baojishan, Gansu	3.47	2	J_{1-2}	Ren et al. [39]	Hengqu, Shanxi	11.87	3	E–N	Tang et al. [66]
Yaojie, Gansu	3	1	J_{1-2}	Ren et al. [39]	Shanxi	1.37	57	C–P	Tang et al. [66]
Wangjiashan, Gansu	2	1	J_{1-2}	Ren et al. [39]	Hunyuan, Shanxi	4.7	1	C–P	Zhang et al. [107]
Guangzhou, Guangdong	13.9	1	P_2	Lu et al. [79]	Pingshuo, Shanxi	6.9	8	C–P	Zhuang et al. [128]
Xingmei, Guangdong	8	1	P_2	Ren et al. [39]	Pingshuo, Shanxi	3.9	2	C–P	Zhao et al. [89]
Guangdong	9.6	33		Cui and Chen [80]	Shuozhou Pinglu, Shanxi	0.2	1	C–P	Zhang et al. [107]
Baise, Guangxi	25.4	2	E–N	Ren et al. [39]	Zuoquan, Shanxi	6.8	1	C–P	Zhang et al. [107]
Nanning, Guangxi	19.6	2	E–N	Ren et al. [39]	Yangquan, Shanxi	0.7	1	C–P	Zhang et al. [107]
Hongmao, Guangxi	3.84	1	C–P	Ren et al. [39]	Yangquan, Shanxi	1.23	2	C–P	Zhao et al. [89]
Heshan, Guangxi	3.36	12	P_2	Li et al. [81]	Lingshi, Shanxi	1	1	C–P	Zhang et al. [107]
Fusui, Guangxi	8.59	10	P_2	Dai et al. [82]	Xishan, Shanxi	3.4	1	C–P	Tang et al. [66]
Heshan, Guangxi	13.58	12	P_2	Shao et al. [83]	Fenxi, Shanxi	0.8	3	C–P	Zhang et al. [107]
Heshan, Guangxi	11.8	4	P_2	Dai et al. [84]	Huoxi, Shanxi	2	7	C–P	Tang et al. [66]
Heshan, Guangxi	3.37	6	P_2	Dai et al. [84]	Pingshuo, Shanxi	2.96	6	C–P	Zhao [30]
Heshan, Guangxi	5.31	2	P_2	Dai et al. [84]	Pingshuo, Shanxi	0.4	57	C–P	Ren et al. [39]
Yishan, Guangxi	8.3	22	P_3	Dai et al. [85]	Hedong, Shanxi	0.99	29	C–P	Li [111]
Shuicheng, Guizhou	0.92	3	P_2	Zen et al. [86]	Huozhou, Shanxi	1.81	9	C–P	Chen and Tang [72]
Shuicheng, Guizhou	6.26	3	P_2	Zen et al. [86]	Xishan, Shanxi	4.27	19	C–P	Ge [129]
Liuzhi Shuicheng, Guizhou	8	45	P_2	Zhuang et al. [87]	Yangquan, Shanxi	1.17	7	C–P	Zhao [30]
Lindong, Guizhou	7.6	32	P_2	Ni et al. [88]	Jincheng, Shanxi	2.22	5	C–P	Wang [130]
Nayong, Guizhou	1.26	1	P_2	Zhao et al. [89]	Lu'an, Shanxi	0.95	1	C–P	Bai [118]
Zhijin Shuchang, Guizhou	26.5	4	P_2	An et al. [90]	Ningwu, Shanxi	0.29	1	C–P	Ren et al. [39]
Zhijin Xingzhai, Guizhou	2.5	3	P_2	An et al. [90]	Xuangang, Shanxi	3.43	1	C–P	Ren et al. [39]
Xingren Jiaole, Guizhou	10.8	18	P_2	Zhou et al. [91]	Datong, Shanxi	8	6	C–P	Ren et al. [39]
Dafang, Guizhou	5.79	71	P_2	Dai et al. [92]	Hongdong, Shanxi	2.87	5	C–P	Ren et al. [39]
Panxian, Guizhou	3.68	16	P_2	Feng et al. [93]	Changzhi, Shanxi	2	1	C–P	Ren et al. [39]
Shuicheng, Guizhou	7.15	25	P_2	Feng et al. [93]	Gujiao, Shanxi	1.47	3	C–P	Ge [129]
Guiyang, Guizhou	7.27	3	P_2	Feng et al. [93]	Gujiao, Shanxi	6.73	2	C–P	Ge [129]
Liuzhi, Guizhou	8.52	15	P_2	Feng et al. [93]	Pingshuo, Shanxi	3.91	2	C–P	Zhao et al. [89]
Nayong, Guizhou	2.59	6	P_2	Zhou [31]	Fenxi, Shanxi	1.1	1	C–P	Zhang et al. [107]
Zhijin, Guizhou	5.27	59	P_2	Zhou [31]	Xishan, Shanxi	0.75	2	C–P	Zhang et al. [107]
Zhijin, Guizhou	4.88	15	P_2	Dai et al. [38]	Shanxi	2.07	89	C–P	Zhang et al. [131]
Zhijin, Guizhou	1.1	1	P_2	Dai et al. [37]	Fanshi and Yuanqu, Shanxi	13.5	4	P_3	Zhang et al. [131]
Qinglong, Guizhou	39.15	4	P_2	Zhang [63]	Datong, Shanxi	7.61	30	J_{1-2}	Zhuang et al. [128]
Xishui, Guizhou	3.19	7	P_2	Ren et al. [39]	Datong, Shanxi	8.5	3	J_{1-2}	Zhang et al. [107]
Bijie, Guizhou	5.54	4	P_2	Dai et al. [94]	Datong, Shanxi	4.79	8	J_{1-2}	Zhuang et al. [100]
Guiding, Guizhou	6.06	1	P_2	Lei [95]	Datong, Shanxi	3.84	8	J_{1-2}	Chen et al. [71]
southwest of Guizhou	10.72	36	P_2	Zhang [63]	Datong, Shanxi	12.3	17	J_{1-2}	Zhang et al. [131]
Guizhou	12.51	1	P_2	Luo et al. [75]	Pubai, Shaanxi	2.14	1	C–P	Ren et al. [39]
Qinglong, Guizhou	1.58	15	P_2	Li and Tang [96]	Chenghe, Shaanxi	1.36	1	C–P	Ren et al. [39]

Table 1. *Cont.*

Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source	Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source
Liupanshui, Guizhou	5.87	62	P_2	Guo et al. [97]	Hancheng, Shaanxi	1	1	C–P	Ren et al. [39]
Xingren, Zhijin, Liuzhi, Bijie, Dafang, Guizhou	3.9	71	P_2	Dai et al. [94]	Tongchuan, Shaanxi	2.43	2	C–P	Ren et al. [39]
Guiding, Guizhou	9.24	14	P_2	Dai et al. [98]	Weibei, Shaanxi	7.595	48	C–P	Lu [132]
Zhenfeng Longtoushan, Guizhou	54.77	7	P_3	Zhang [63]	Hancheng and Tongchuan, Shaanxi	3.88	14	C–P	Wang et al. [133]
Puan, Guizhou	10.18	5	P_2	Yang [99]	Zichang, Shaanxi	1.5	1	P_3	Ren et al. [39]
Changpo, Hainan	8.07	1	E–N	Ren et al. [39]	Binxian, Shaanxi	13.64	1	J_{1-2}	Li et al. [119]
Tangshan Jinggezhuang, Hebei	9.52	1	C–P	Zhuang et al. [100]	Junlian, Sichuan	26.5	1	P_2	Zhang et al. [107]
Kailuan, Hebei	6.14	47	C–P	Tang et al. [101]	Guxu, Sichuan	4.93	30	P_2	Ren et al. [39]
Kailuan, Hebei	6.41	34	C–P	Zhuang et al. [100]	Junlian, Sichuan	10.65	6	P_2	Ren et al. [39]
Fengfeng, Hebei	2.38	9	C–P	Dai [102]	Furong, Sichuan	9.48	5	P_2	Ren et al. [39]
Handan, Hebei	3.83	3	C–P	Ren et al. [39]	Dabaoding, Sichuan	1.44	31	P_2	this study
Xingtai, Hebei	1.6	3	C–P	Ren et al. [39]	Huayingshan, Sichuan	3.15	5	P_2	Dai et al. [134]
JingXing and Yuanshi, Hebei	2.28	2	C–P	Ren et al. [39]	Dukou, Sichuan	1.99	2	P_3	Ren et al. [39]
Xinglong, Hebei	3.53	1	C–P	Ren et al. [39]	Yaxing, Sichuan	3.27	5	P_3	Ren et al. [39]
Fengfeng, Hebei	15.03	15	P_2	Dai and Ren [103]	Guxu, Sichuan	2.252	11	P_2	Dai et al. [135]
Jiaozuo, Henan	1.02	2	C–P	Ren et al. [39]	Taiwan	25.38	4	E–N	Ren et al. [39]
Pingdingshan, Henan	1.93	22	C–P	Chen and Tang [72]	Tibet	20.11	24	P_3	Fu et al. [136]
Hebi, Henan	1.56	2	C–P	Zhao [30]	Kuche, Xinjiang	0.85	15	J_{1-2}	Chen and Tang [72]
Xinmi, Henan	1.13	1	C–P	Ren et al. [39]	Yining, Xinjiang	1.85	10	J_{1-2}	Chen and Tang [72]
Yongcheng, Henan	2	1	C–P	Ren et al. [39]	Santanghu, Xinjiang	3.86	4	J_{1-2}	Chen and Tang [72]
Yima, Henan	19.44	5	J_{1-2}	Ren et al. [39]	Liuhuanggou, Xinjiang	1.49	5	J_{1-2}	Chen and Tang [72]
Jiayin, Heilongjiang	2.77	1	E–N	Ren et al. [39]	Fukang, Xinjiang	4.89	5	J_{1-2}	Chen and Tang [72]
Jixi, Heilongjiang	1.95	7	J_3–K_1	Ren et al. [39]	Lucaogou, Xinjiang	5.55	1	J_{1-2}	Ren et al. [39]
Shuangyashan, Heilongjiang	17.2	6	J_3–K_1	Ren et al. [39]	Aiweiergou, Xinjiang	0.96	10	J_{1-2}	Chen and Tang [72]
Qitaihe, Heilongjiang	1.85	3	J_3–K_1	Ren et al. [39]	Hami, Xinjiang	1.98	13	J_{1-2}	Chen and Tang [72]
Suibin, Heilongjiang	8.75	4	J_3–K_1	Ren et al. [39]	Miquan, Xinjiang	10.25	1	J_{1-2}	Ren et al. [39]
Jixian, Heilongjiang	4.47	8	J_3–K_1	Ren et al. [39]	Dapugou, Xinjiang	4.69	1	J_{1-2}	Ren et al. [39]
Hegang, Heilongjiang	1.67	1	J_3–K_1	Ren et al. [39]	Yili, Xinjiang	20.027	40	J_{1-2}	Dai et al. [137]
Heihe, Heilongjiang	2.85	1	J_3–K_1	Ren et al. [39]	Hami, Xinjiang	2.13	10	J_{1-2}	Tang et al. [66]
Dazhi, Hubei	17.26	3	P_2	Ren et al. [39]	Fukang, Xinjiang	1.24	4	J_{1-2}	Tang et al. [66]
Songyi, Hubei	4.58	1	P_2	Ren et al. [39]	Aiweiergou, Xinjiang	0.58	5	J_{1-2}	Tang et al. [66]
Lichuan, Hubei	8.3	1	P_2	Ren et al. [39]	Kuche, Xinjiang	0.59	6	J_{1-2}	Tang et al. [66]
Hubei	5.1	19		Cui and Chen [80]	Hetian, Xinjiang	1.76	1	J_{1-2}	Tang et al. [66]
Lianshao Jinzhushan, Hunan	2	1	C–P	Ren et al. [39]	Zhunnan, Xinjiang	1.49	5	J_{1-2}	Tang et al. [66]
Lianshao Lengshuijiang, Hunan	23	1	C–P	Ren et al. [39]	Kuche, Xinjiang	2.18	9	J_{1-2}	Tang et al. [66]
Zhadu, Hunan	4.14	17	C–P	Yuan [104]	Lincang bangmai, Yunnan	10.8	1	E–N	Tang et al. [66]
Meitian, Hunan	10	10	P_2	Tang et al. [66]	Lincang, Yunnan	12	4	E–N	Tang et al. [66]
Lianshao, Hunan	2.4	3	P_2	Ren et al. [39]	Zhaotong, Yunnan	9.75	25	E–N	Ren et al. [39]
Baisha, Hunan	4.75	1	P_2	Ren et al. [39]	Xiaolongtan, Yunnan	19.92	8	E–N	Ren et al. [39]
Meitanba, Hunan	6.05	1	P_2	Ren et al. [39]	Xianfeng, Yunnan	2.88	16	E–N	Ren et al. [39]
Zixing, Hunan	23.29	2	P_3	Ren et al. [39]	Jinsuo, Yunnan	7.01	2	E–N	Gu [138]

Table 1. *Cont.*

Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source	Province/Coal Mine	Arithmetic Mean	Sample Number	Coal-Forming Period	Data Source
Meihe, Jilin	6	1	E–N	Ren et al. [39]	Kebao, Yunnan	5.09	19	E–N	Ren et al. [39]
Shulan, Jilin	2.51	3	E–N	Ren et al. [39]	Fengmingcun, Yunnan	2.5	1	E–N	Ren et al. [39]
Baishan, Jilin	7.45	56	C–P	Wu et al. [105]	Longling Daba, Yunnan	6.98	2	E–N	Ren et al. [39]
Liaoyuan, Jilin	21	1	J_{1-2}	Ren et al. [39]	Chuxiong, Yunnan	17.39	3	E–N	Ren et al. [39]
Xuzhou Chacheng, Jiangsu	1.1	7	C–P	Tang et al. [66]	Yaoan, Yunnan	6.43	184	E–N	Ren et al. [39]
Xuzhou, Jiangsu	2.04	12	C–P	Chen and Tang [72]	Kunming, Yunnan	25.8	23	E–N	Ren et al. [39]
Fengpei, Jiangsu	3.88	5	C–P	Ren et al. [39]	Yuxi, Yunnan	30.5	14	E–N	Zhou [139]
Zhenjiang, Jiangsu	1.5	2	P_2	Ren et al. [39]	Zhaotong, Yunnan	12.7	24	E–N	Zhou [139]
Ganzhong, Jiangxi	11.87	240	P_2	Zhou [106]	Wandian, Yunnan	21.4	33	E–N	Zhou [139]
Leping, Jiangxi	9.5	1	P_2	Zhang et al. [107]	Baolang, Yunnan	7	17	E–N	Zhou [139]
Fengcheng, Jiangxi	9.5	70	P_2	Zhou [106]	Yaoan, Yunnan	8.5	126	E–N	Zhou [139]
Yinggangling, Jiangxi	10.9	192	P_2	Zhou [106]	Lincang, Yunnan	47.6	52	E–N	Dai et al. [40]
Yangqiao, Jiangxi	4.18	21	P_2	Zhou [106]	Lincang, Yunnan	117	11	E–N	Dai et al. [62]
Pingxiang, Jiangxi	1.65	8	P_3	Ren et al. [39]	Yanshan, Yunnan	9.1	3	P_2	Dai et al. [140]
Luoshi, Jiangxi	3.1	70	P_3	Zhou [106]	Laochang, Yunnan	5	175	P_2	Zhou [31]
Huagushan, Jiangxi	3.28	8	P_3	Zhou [106]	Enhong, Yunnan	1.77	55	P_2	Zhou [31]
Yongshan, Jiangxi	11	7	P_3	Ren et al. [39]	Yangchang, Yunnan	0.9	97	P_2	Zhou [31]
Ganzhong, Jiangxi	3.12	78	P_3	Zhou [106]	Laibin, Yunnan	1.29	12	P_2	Zhou [31]
Shenbei, Liaoning	9.88	7	E–N	Ren et al. [108]	Housuo and Qingyun, Yunnan	0.63	2	P_2	Ren et al. [39]
Fushunxi, Liaoning	2.18	1	E–N	Tang et al. [66]	Qujing, Yunnan	1.13	1	P_2	Ren et al. [39]
Fushun, Liaoning	3.36	2	E–N	Ren et al. [39]	Luoping, Yunnan	4.88	1	P_2	Ren et al. [39]
Shenbei, Liaoning	16.85	12	E–N	Ren et al. [39]	Yanshan, Yunnan	15.5	1	P_2	Ren et al. [39]
Tiefa, Liaoning	4.1	3	J_3–K_1	Ren et al. [39]	Xuanwei, Yunnan	8.37	6	P_2	Dai et al. [141]
Fuxin, Liaoning	4.39	7	J_3–K_1	Zhuang et al. [100]	Yanshan, Yunnan	9.14	6	P_2	Dai et al. [142]
Badaohao, Liaoning	23.3	1	J_3–K_1	Ren et al. [39]	Yipinglang, Yunnan	21.85	2	P_3	Ren et al. [39]
Fuxin Haizhou, Liaoning	4.98	6	J_3–K_1	Querol et al. [109]	Chuxiong, Yunnan	11.4	1	P_3	Ren et al. [39]
Beipiao, Liaoning	3.73	1	J_{1-2}	Ren et al. [39]	Xinping, Yunnan	4.6	1	P_3	Ren et al. [39]
Gongwusu, Inner Mongolia	1.2	1	C–P	Wang et al. [110]	Xinde, Yunnan	2.53	4	P_2	Dai et al. [143]
Wuda, Inner Mongolia	1.57	4	C–P	Dai [102]	Xinde, Yunnan	1.14	3	P_2	Dai et al. [143]
Jungar, Inner Mongolia	0.85	3	C–P	Li [111]	Luquan, Yunnan	5.28	8	D	Dai et al. [144]
Wuhai, Inner Mongolia	3.96	1	C–P	Ren et al. [39]	Zhejiang	11	5		Cui and Chen [80]
Daqingshan, Inner Mongolia	1.51	33	C–P	Dai et al. [112]	Changguang, Zhejiang	13	8	P_2	Ren et al. [39]

Table 2. Arsenic concentrations and predicted coal resources in individual provinces of China.

Administrative Division	Predicted Resource/Billion Tons [64]	Sample Number	Arsenic Content Mean Value (mg/kg)	Concentration Coefficient
Anhui	611.59	350	6.32	1.67
Beijing	86.72	4	1.47	0.39
Fujian	25.57	5	10.20	2.69
Gansu	1428.87	14	2.86	0.75
Guangdong	9.11	35	9.68	2.55
Guangxi	17.64	73	8.82	2.33
Guizhou	1896.9	547	7.25	1.91
Hainan	0.01	1	8.07	2.13
Hebei	601.39	115	6.85	1.81
Henan	919.71	33	4.48	1.18
Heilongjiang	176.13	31	6.47	1.71
Hubei	2.04	24	6.73	1.78
Hunan	45.35	36	7.22	1.91
Jilin	30.03	61	7.41	1.96
Jiangsu	50.49	26	2.10	0.55
Jiangxi	40.84	695	9.04	2.39
Liaoning	59.27	40	9.51	2.51
Inner Mongolia	12,250.4	1053	3.80	1.00
Ningxia	1721.11	30	1.72	0.45
Qinghai	380.42	20	1.17	0.31
Shandong	405.13	250	7.49	1.98
Shanxi	3899.18	404	3.13	0.83
Shaanxi	2031.1	69	6.43	1.70
Sichuan and Chongqing	303.79	269	5.38	1.42
Taiwan	1.8	4	25.38	6.70
Tibet	8.09	24	20.11	5.31
Xinjiang	18,037.3	145	6.88	1.82
Yunnan	437.87	943	10.82	2.85
Zhejiang	0.44	13	12.23	3.23
China	45,478.3	5314	6.97 5.33 (weighted average)	

Table 3. Arsenic content in Chinese coals (mg/kg). Data are quoted from references of [3,19,21,23,39–41,44,45,47,56,57,66,145].

Country	Arithmetic Mean	Geometric Mean	Resource Weighted Average	Sample Number	Data Source
Chinese Coal	276.61	4.26		132	Ren et al. [56]
Chinese Coal	4.7			1018	Wang [145]
Chinese Coal	4			1915	Chen et al. [57]
Chinese Coal	5			3193	Tang et al. [66]
Chinese Coal	6.4	3.96		297	Wang [23]
Chinese Coal			3.79	3386	Dai et al. [40]
Chinese Coal			3.80	3453	Ren et al. [39]
Chinese Coal	9.70		3.18	4805	Kang [3]
Chinese Coal	6.97		5.33	5314	this study
American Coal	24	6.5		7676	Finkelman [41]
American Coal	24			6878	Kolker et al. [21]
Australian Coal	2				Swaine and Goodarzi [44]
Czech Coal	39.94			9172	Bouska and Pesek [45]
World lignite	7.6 ± 1.3			21,092	Ketris and Yudovich [19]
World bitumite	9.0 ± 0.7			22,466	Ketris and Yudovich [19]
World Coal	5				PECH [47]

3. Distribution of Arsenic in Chinese Coals

3.1. Spatial Distribution Characteristics of Arsenic in Chinese Coals

Based on the collected data of arsenic content in Chinese coals, the average arsenic contents in coals from different coal-bearing areas are calculated. Additionally, compared to average value of Chinese coals reported by Dai et al. [40], the concentration coefficient (CC) is given in Table 4. When CC is above 100, it is abnormally enriched; when CC is above 10 but below 100, it is highly enriched; when CC is above 5 but below 10, it is enriched; when CC is above 2 but below 5, it is slightly enriched; when CC is above 0.5 but below 2, it is normal; and when CC is below 0.5, it is depleted [112].

In view of the concentration coefficient, the spatial variation of arsenic in Chinese coals can be classified as follow:

(1) Arsenic in coal is highly enriched in the southwestern Yunnan and part of Guizhou.
(2) Arsenic is enriched in the coals from some regions, such as the western Yunnan, Guangxi, Taiwan, Tibet, southwestern Liaoning, Jilin, and Henan.
(3) Arsenic in coal is slightly enriched in the southwestern and middle part of Guizhou, most of Yunnan, Guangdong, Guangxi, Hainan, Fujian, Jiangxi, Zhejiang, southeastern Hubei, southern Hunan, northwestern Chongqing, southwestern Shandong, southern Hebei, southern Shanxi, as well as parts of northern and southern Liaoning.
(4) Arsenic in coal is depleted in the western Xinjiang and most of Qinghai.

Overall, the content of arsenic in Chinese coals has an increasing tendency from the north to the south. Meanwhile, the content of arsenic in coal within coal-bearing basins differs spatially, due to factors, such as palaeomire conditions and provenance supply. The arsenic contents in coals from different provinces are various obviously. The arsenic content in coal has a significant correlation with coal-accumulation area. The distribution of arsenic in nationwide China is shown in Figure 1. The population of coal samples from Taiwan, Beijng, and Fujian are small and cannot well-represented local coals.

Table 4. Spatial variation of arsenic in Chinese coals.

Province/Area	**Anhui Province**	**Northern Anhui**	**Southern Anhui**	**North Central of Anhui**	**Beijing**	**Western Beijing**	**Fujian Province**	**Central of Fujian**
Mean Value **CC**	6.32 1.67	2.35 0.62	10.16 2.68	6.37 1.68	1.47 0.39	1.00 0.26	10.20 2.69	12.00 3.17
Province/Area	Gansu Province	Eastern Gansu	Southeastern Gansu	Northwestern Gansu	Central of Gansu	Guangdong Province	Northeastern Guangdong	Southern Guangdong
Mean Value **CC**	2.86 0.75	3.33 0.88	1.50 0.40	3.06 0.81	2.99 0.79	9.68 2.55	8.00 2.11	13.90 3.67
Province/Area	Guangxi	Northern Guangxi	Southern Guangxi	Western Guangxi	Southwestern Guangxi	Central of Guangxi	Guizhou Province	Northern Guizhou
Mean Value **CC**	8.82 2.33	3.84 1.01	19.60 5.17	25.40 6.70	8.59 2.27	7.81 2.06	7.25 1.91	3.19 0.84
Province/Area	Northwestern Guizhou	Western Guizhou	Southwestern Guizhou	Central of Guizhou	Hainan Province	Northwestern Hainan	Hebei Province	Northeastern Hebei
Mean Value **CC**	5.78 1.52	5.91 1.56	12.41 3.28	8.01 2.11	8.07 2.13	8.07 2.13	6.85 1.81	3.53 0.93
Province/Area	Eastern Hebei	Southern Hebei	Western Hebei	Henan Province	Northern Henan	Eastern Henan	Western Henan	Central of Henan
Mean Value **CC**	6.29 1.66	8.77 2.31	2.28 0.60	4.48 1.18	1.29 0.34	2.00 0.53	19.44 5.13	1.90 0.50
Province/Area	Heilongjiang Province	Northern Heilongjiang	Northeastern Heilongjiang	Eastern Heilongjiang	Southeastern Heilongjiang	Northwestern Heilongjiang	Hubei Province	Southeastern Hubei
Mean Value **CC**	6.47 1.71	2.77 0.73	9.24 2.44	1.85 0.49	1.95 0.51	2.85 0.75	6.73 1.78	17.26 4.55
Province/Area	Southwestern Hubei	Hunan Province	Southern Hunan	Central of Hunan	Jilin Province	Northern Jilin	Southern Jilin	South Central of Jilin
Mean Value **CC**	6.44 1.70	7.22 1.91	11.64 3.07	4.72 1.25	7.41 1.95	2.51 0.66	7.42 1.96	21.00 5.54
Province/Area	Jiangsu Province	Northwestern Jiangsu	Southwestern Jiangsu	Jiangxi Province	Northeastern Jiangxi	Western Jiangxi	Central of Jiangxi	Liaoning Province
Mean Value **CC**	2.10 0.55	2.15 0.57	1.50 0.40	9.04 2.38	10.81 2.85	8.15 2.15	9.68 2.55	9.51 2.51
Province/Area	Northern Liaoning	Eastern Liaoning	Western Liaoning	Southwestern Liaoning	Inner Mongolia(IM)	Northeastern IM	Southeastern IM	Western IM

Table 4. *Cont.*

Province/Area	**Anhui Province**	**Northern Anhui**	**Southern Anhui**	**North Central of Anhui**	**Beijing**	**Western Beijing**	**Fujian Province**	**Central of Fujian**
Mean Value **CC**	12.89 3.40	2.97 0.78	4.60 1.21	23.30 6.15	3.80 1.00	7.83 2.07	2.95 0.78	2.59 0.68
Province/Area	Southwestern IM	Midwestern IM	Ningxia	Northern Ningxia	Central of Ningxia	Qinghai Province	Northern Qinghai	Eastern Qinghai
Mean Value **CC**	3.76 0.99	1.39 0.37	1.72 0.45	1.12 0.30	4.13 1.09	1.17 0.31	0.90 0.24	3.34 0.88
Province/Area	Western Qinghai	Shandong Province	Northeastern Shandong	Southern Shandong	Western Shandong	Southwestern Shandong	Central of Shandong	Shanxi Province
Mean Value **CC**	1.34 0.35	7.49 1.98	2.90 0.77	5.23 1.38	7.90 2.08	8.78 2.32	6.12 1.61	3.13 0.83
Province/Area	Northern Shanxi	Northeastern Shanxi	Eastern Shanxi	Southeastern Shanxi	Southern Shanxi	Northwestern Shanxi	Southwestern Shanxi	Central of Shanxi
Mean Value **CC**	7.89 2.08	10.57 2.79	1.14 0.30	2.01 0.53	7.70 2.03	1.47 0.39	1.31 0.35	3.49 0.92
Province/Area	Shaanxi Province	Northern Shaanxi	Central of Shaanxi	Sichuan Province	Northeastern Sichuan	Eastern Sichuan	Southeastern Sichuan	Southwestern Sichuan
Mean Value **CC**	6.43 1.70	1.50 0.40	6.50 1.71	4.07 1.07	1.99 0.53	3.15 0.83	5.86 1.55	1.44 0.38
Province/Area	Central of Sichuan	Taiwan	Tibet	Xinjiang	Northern Xinjiang	Eastern Xinjiang	Northwestern Xinjiang	Southwestern Xinjiang
Mean Value **CC**	3.27 0.86	25.38 6.70	20.11 5.31	6.88 1.81	2.63 0.69	2.31 0.61	16.39 4.32	1.76 0.46
Province/Area	North Central of Xinjiang	Midwest of Xinjiang	Yunnan Province	Northeastern Yunnan	Eastern Yunnan	Southeastern Yunnan	Western Yunnan	Southwestern Yunnan
Mean Value **CC**	1.76 0.46	1.20 0.32	10.82 2.85	9.03 2.38	3.13 0.83	14.28 3.77	20.58 5.43	56.19 14.83
Province/Area	Central of Yunnan	Zhejiang Province	Northern Zhejiang	Chongqing	Northern Chongqing	Southeastern Chongqing	Southern Chongqing	Western Chongqing
Mean Value **CC**	9.14 2.41	12.23 3.23	13.00 3.43	6.11 1.61	7.88 2.08	7.40 1.95	4.96 1.31	8.77 2.31

Concentration coefficient short for CC.

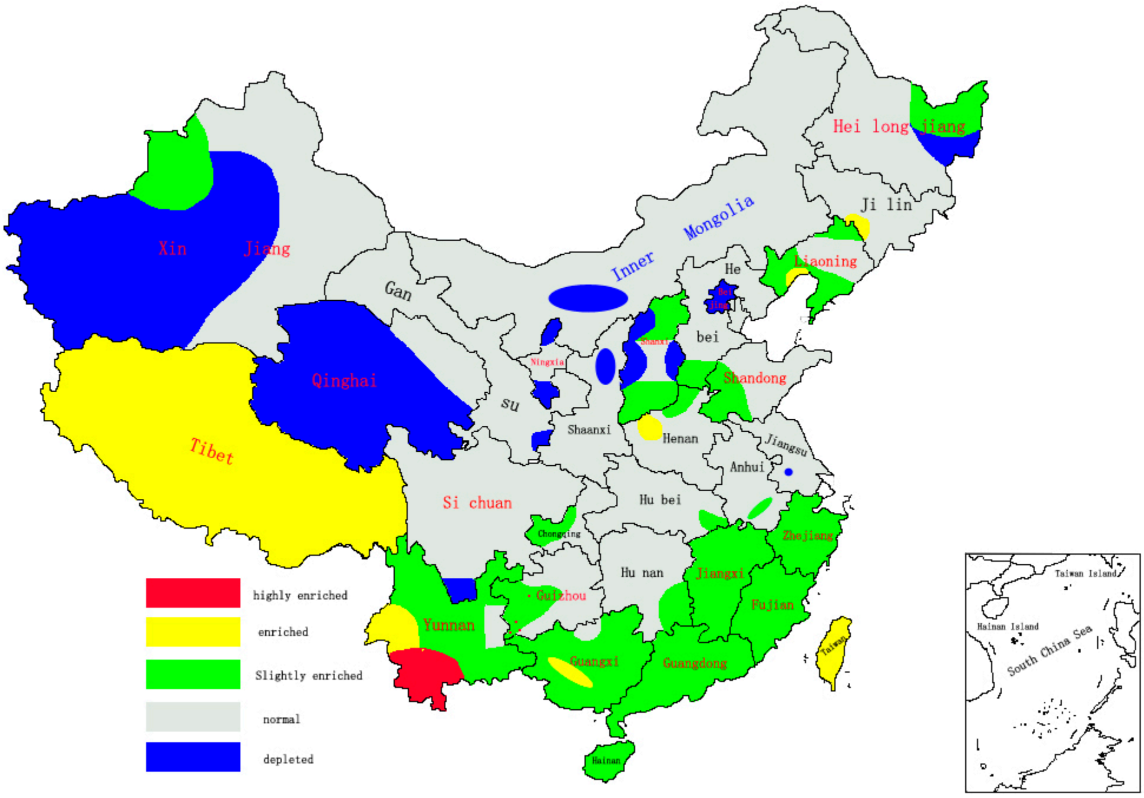

Figure 1. Arsenic distribution in Chinese coals by provinces.

3.2. Distribution Characteristic of Arsenic in Chinese Coal in Different Coal-Forming Periods

There are six major coal-forming periods in China: Late Carboniferous and Early Permian (C_2–P_1), Late Permian (P_2), Late Triassic (T_3), Early and Middle Jurassic (J_{1-2}), Late Jurassic and Early Cretaceous (J_3–K_1), and Paleogene and Neogene (E–N) [39]. Coals of these periods individually account for 38.1%, 7.5%, 0.4%, 39.6%, 12.1%, and 2.3% of the total Chinese reserves based on the Third National Prediction of Coal Resources of China [40].

The arsenic contents in coals of various coal-forming periods exhibit a significant difference [146]. The calculated contents of arsenic in coals shown in Table 5 exhibit the following regularity: Paleogene and Neogene > Late Triassic > Late Permian > Late Jurassic and Early Cretaceous > Early and Middle Jurassic > Late Carboniferous and Early Permian, which is similar to the trend reported by Wang [23]. However, it differs from the distribution reported by Zheng et al. [146] and Lv et al. [147], who considered the averaging arsenic content in the Triassic coals was at first. The content of arsenic in low rank coals is highest in the Paleogene and Neogene, which is consistent with the results of Zhou [139], Li et al. [148], and Dai et al. [62]. Due to the great number of samples and an elaborative analysis and verification of the data sources, this statistical conclusion might be credible.

Table 5. Arsenic concentration in coals of different coal-forming periods in China (mg/kg).

Coal-Forming Periods	Arithmetic Mean	Sample Number	Recoverable Coal Reserve (%)
Late Carboniferous and Early Permian	4.63	1316	38.1
Late Permian	7.13	1839	7.5
Late Triassic	7.76	257	0.4
Early and Middle Jurassic	4.66	1141	39.6
Late Jurassic and Early Cretaceous	6.88	88	12.1
Paleogene and Neogene	15.5	606	2.3

3.3. Profile Distribution of Arsenic in Chinese Coals

From a profile distribution perspective, there are obvious variations of arsenic content in different coal seams. Generally, arsenic variation in the coal-bearing profiles can be divided into several types: (1) Arsenic enriches in the roof, floor, coal seam and parting materials, e.g., the Xinlongchang and Jiaole coal mines in the Xingren of Guizhou Province [149]; (2) Arsenic enriches in the roof while depletes in the coal seam, e.g., the No. 15 coal of Qinshui Basin [150], the Nos. 4 and 6 coals of the Donglin coal mine in Nantong coalfield of Chongqing [151], and the No. 24 coal of the Taiping coal mine in the Panzhihua; (3) Arsenic enriches in the floor while depletes in the coal seam, e.g., the No. 3 coal of the Qinshui Basin [150] and the Xiashan coal mine in the Xingren of Guizhou Province [149]; (4) Arsenic in both the roof and floor are enriched, e.g., the Taiyang coal mine in the middle of Jiangxi Province [106]; And (5) arsenic content presents no obvious variation in one coal seam, e.g., the No. 5 coal of Chuancaogedan in Jungar Coalfield [114]. In a thick, multi-layer coal seam of the Panjiazhuang coal mine in the Xinren of Guizhou Province, only one layer contains high concentrations of As [149].

4. Modes of Occurrence of Arsenic in Chinese Coals

The modes of occurrence of arsenic in coals are of significance in the understanding of arsenic accumulation, migration mechanism, proper utilization of coal resources, and decrease of environment problems [23]. Arsenic in coals can be classified into the inorganic and organic arsenic. The relationship between minerals (such as pyrite, marcasite, and clay minerals) and arsenic in coals were widely investigated [30,117,152–154]. Although the detailed structure of organic arsenic in coal is still uncertain, many researchers [28,32,152,155] have a positive view on the existence of organic arsenic. The modes of occurrence of arsenic in Chinese coals include sulfide-association, organic-association, arsenate-association, silicate-association, and soluble- and exchangeable-association.

4.1. Sulfide-Association

Arsenic in coal usually co-exists with pyrite [28,152–154], but it is rare to find arsenic in the form of realgar and orpiment. Zhou [31] researched anthracite coal in the Late Permian Laochang mining area in the eastern Yunnan and discovered that the pyrite was the main carrier of arsenic when sulfur contents higher than 0.6%. By sequential extraction, Guo et al. [156] measured the occurrence of arsenic in anthracite, lignite and bituminous coal, indicating that 73%–83% arsenic was bound to sulfide. Moreover, Zhao et al. [152] found that sulfide-bound arsenic, as pyrite, accounts for 0–85%, with an average of 36%. Arsenic in pyrite in coal exists mainly in the form of arsenic-bearing pyrite, rather than arsenopyrite. By electronic probe analysis on a high arsenic coal in the western Guizhou, Nie and Xie [157] confirmed that arsenic existed mainly in pyrite, and with distinctly different arsenic content on the high side of the secondary pyrite. The results of Zhao [30] and Zhang [63] on the arsenic in pyrite of the Late Permian in Guizhou suggested that the arsenic in epigenetic low-temperature hydrothermal vein pyrite was higher than that of syngenetic pyrite. The arsenic enrichment in Guizhou coals resulted from the epigenetic low-temperature hydrothermal fluids. Chen et al. [151] also found that arsenic primarily associated with pyrite in the Donglin coal from Nantong coalfield of Chongqing.

4.2. Organic-Association

Organic-associated arsenic is ubiquitous in coal and the proportion of organic-associated arsenic varies considerably among different coal samples. Using an extraction experiment of arsenic in low rank Xiaolongtan coals from the Yunnan Province, Zhang and Fan [155] discovered that more than 80% of total arsenic was organic associated. Wang et al. [158] found that organic-associated arsenic accounted for 51.38%–100% in the Jincheng coal from Shanxi Province. Zhao et al. [89] concluded that when the content of arsenic in coal was below 5.5 mg/kg and ash content was below 30%, arsenic presented as organic-association. About 8% arsenic in the Laiyang anthracite coal, Qianjiaying lignite, and Qingshan bituminous coal was organic-associated [156]. Liu et al. [159] and Ding et al. [160]

focused on the Yanzhou coal and the high-arsenic coal in the southwestern Guizhou, respectively, an organic-association arsenic was reported.

4.3. Arsenate-Association

Leaching experiments on lignites from Yunnan indicate that besides the arsenate absorbed by iron oxide and hydroxide, the arsenic mainly exists in limonite, magnetite, hematite, and other iron minerals [138]. Ren et al. [56] also found that arsenic occurred as arsenate in coals. Zhao et al. [59] and Ding et al. [32] investigated the Late Permian high-arsenic coal from Guizhou Province, arsenic were found in forms of arsenate or arsenite. Further, Zhao et al. [152] reported that arsenic combined with arsenate in coals accounts for approximately 0–65%, with 17% on average. The proportion of arsenate-state arsenic is positively correlated with the iron content in coal [152].

4.4. Silicate-Association

Chen et al. [57] found that the arsenic presents an increasing trend with the increase of Al_2O_3 and Fe_2O_3. And they inferred that the clay minerals contain arsenic. By the sequential chemical extraction experiments on some samples from the Guizhou and Shanxi, Zhao et al. [152] found that silicate-combined arsenic accounts for approximately 0–90% of total arsenic, with an average of 16%, which is proportional to the logarithm of ash yield in coal. The silicate-associated arsenic that is extracted from coal by hydrofluoric acid is mainly in the crystal lattice of clay minerals.

4.5. Soluble- and Exchangeable-Association

Arsenic in soluble and exchangeable forms is easily mobilized, and thus, have an adverse impact on the environment and human health. In a Huainan coal, Kang [3] reported that the ratio of the total soluble to exchangeable arsenic is between 1.78% and 6.28%, illustrating that the arsenic in coal has certain dissolution ability and could be released into the surface environment by rainfall.

4.6. Summary of Modes of Occurrence of Arsenic in Chinese Coals

Based on the sequential chemical extraction experiments on high-arsenic coal in the Guizhou, Ding [161] found that organic-associated arsenic accounts for 0–80%, silicate-associated arsenic accounts for 15%–90%, sulfide-associated arsenic accounts for 0–25%, and arsenate-associated arsenic accounts for 5%–65%. Kang [3] indicated that the modes of occurrence of arsenic in the Huainan coals are mainly sulfide-associated, partially in organic and silicate combined states. Zhao et al. [152] provided a sequence of the range of arsenic modes of occurrence in coals: sulfide-associated (36%) > organic-associated (26%) > arsenate-associated (17%) > silicate-associated (16%) > soluble- and exchangeable-associated (5%). The arsenic in Chinese coals mainly occurs in the form of sulfides-association with considerable differences among the coal samples. Arsenic in As-rich coal is often associated with minerals of epigenetic hydrothermal solution origins.

5. Conclusions

(1) Based on 5314 samples of Chinese coals, the arithmetic mean of the arsenic is calculated as 6.97 mg/kg. Selecting the percentage of coal resources in each province of the total national resources as the weighting factor, the weighted arsenic average is 5.33 mg/kg. Although the arsenic appears abnormally enriched in some Guizhou and Yunnan coals, the common Chinese coal is still comparable to the world level.

(2) The content of arsenic in Chinese coals increases from the north to the south. High-arsenic coal is mainly located in the southwestern Yunnan and part of Guizhou Province. Arsenic is enriched in the coals from some regions, i.e., the western Yunnan, Guangxi, Tibet, southwestern Liaoning, Jilin, and Henan.

(3) The arsenic content in coals of different coal-forming periods shows an overall regularity: Paleogene and Neogene > Late Triassic > Late Permian > Late Jurassic and Early Cretaceous > Early and Middle Jurassic > Late Carboniferous and Early Permian.
(4) The majority of arsenic in Chinese coals exists in arsenic-bearing pyrite. In coal samples with overall low arsenic content, the organic arsenic is dominant.
(5) The distribution and modes of occurrence of arsenic in Chinese coals are impacted by many factors, e.g., the coal-forming material, depositional environment, and epigenetic processes. Thus, the arsenic distribution is nonuniform, and the modes of occurrence exhibit a diversity and complexity. This needs further investigation and assessment by some advanced methods, such as the XAFS, MSXRF, and XANES spectrum.

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