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Review

Concentration and Distribution of Cadmium in Coals of China

Jing Shi [1,2,3], Wenhui Huang [1,2,3,*], Ping Chen [4], Shuheng Tang [1,2,3] and Xiuyan Chen [5]

1 School of Energy Resources, China University of Geosciences (Beijing), Beijing 100083, China; shijing_5577@163.com (J.S.); tangsh@cugb.edu.cn (S.T.)
2 Key Laboratory of Marine Reservoir Evolution and Hydrocarbon Enrichment Mechanism, Ministry of Education, Beijing 100083, China
3 Beijing Key Laboratory of Unconventional Natural Gas Geological Evaluation and Development Engineering, Beijing 100083, China
4 School of Earth and Environment, Anhui University of Science and Technology, Huainan 232001, China; pchen87@163.com
5 Research Institute of Petroleum Exploration & Development, Petrochina, Beijing 100083, China; chenxiuyan1004@petrochina.com.cn
* Correspondence: 1999011250@cugb.edu.cn; Tel.: +86-010-8232-1058

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Abstract: Cadmium is considered an important toxicant of major environmental and occupational concern. It can contaminate water, soil, and the atmosphere through coal mining, beneficiation, combustion, etc. This paper is based on the published literature, especially those data reported during the recent 10 years, including 2999 individual samples from 116 coalfields or mines in 26 provinces in China. The arithmetic mean of cadmium in Chinese coals is 0.43 μg/g. Taking the coal reserves into consideration, the average value of cadmium in coal is estimated as 0.28 μg/g. Cadmium is mostly enriched in the Southern coal-distribution area during the Late Permian. Furthermore, cadmium is highly enriched in Hunan and Chongqing. The modes of occurrence of cadmium in Chinese coals are quite complex. Cadmium in Chinese coals has been found in sulfides, organic matter, silicate minerals, and other minerals. A marine environment may be the most significant factor that influences the cadmium accumulation in coal from the Southern coal-distribution area during the Late Permian. In addition, hydrothermal fluids, source rocks, and volcanic ash have also influenced the content of cadmium in some coalfields in China.

Keywords: cadmium; abundance; geochemistry; coal; China; modes of occurrence

1. Introduction

Among the heavy metals, cadmium is considered an important toxicant of major environmental and occupational concern [1]. Cadmium is extremely toxic to animals even at low contents. The minimal risk levels for acute and chronic inhalation are 0.00003 mg/m^3 and 0.00001 mg/m^3, respectively [2]. Cadmium can be introduced into the body of animals and people via respiratory and digestive tracts [3]. It has a biological half-life in the kidneys between 10 and 30 years. The kidneys, liver, bones, respiratory, and reproductive systems are the main targets of cadmium intoxication [3–5]. The Itai–itai disease caused by cadmium pollution in 1960s in Japan is well known worldwide [6].

China will continue to be one of the largest coal producers and users in the world, moreover coal makes up about 74% of China's total primary energy consumption [7]. Cadmium released from coal through coal mining, beneficiation, combustion, etc. can contaminate water, soil, and the atmosphere [7–9]. It can be abnormally enriched in some coals under certain geologic conditions. Cadmium was discovered in coal mines around China and coals with an elevated concentration of cadmium have been found in many coalfields such as the Chenxi Coalfield in Hunan [10], the Moxinpo Coalfield in Chongqing [11], etc. Consequently, detailed knowledge on the cadmium in coal is crucial to understanding its environmental impact, geological significance, and environmentally-friendly coal utilization. Based on the published literature, especially the data reported during the last 10 years, this paper provides a comprehensive review of cadmium in Chinese coals including its abundance, distribution characteristics, modes of occurrence, and the origins of enrichment.

2. Abundance of Cadmium in Coals

2.1. Abundance of Cadmium in Chinese Coals

China began enhancing its research into hazardous trace elements in coal in the 1980s [12]. The China Coal Research Institute (CCRI) (1980–1990) [13], Bai [13,14], Tang et al. [15], Ren et al. [16,17], and Dai et al. [7] have all calculated the cadmium concentration in Chinese coals respectively (Table 1). In addition, Dai [18] analyzed the cadmium content in coal from the North China Platform. Li [19] measured the concentration of cadmium in coal of Southwest China.

Table 1. Cadmium abundance in Chinese coals according to different authors (unit: μg/g).

Publications	Number of Samples	Mean	Years
CCRI [13]	1018	1.00	1980–1990
Ren et al. [16]	36	0.46	1999
Tang et al. [15]	1307	0.30	2003
Bai [13]	1018	0.91	2003
Ren et al. [17]	1317	0.24	2006
Bai et al. [14]	1123	0.81	2007
Dai et al. [7]	1384	0.25	2012

So far, China has not had a national survey on trace elements in coal. Based on the published literature, 2999 individual samples from 116 coalfields or mines in 26 provinces are presented in this paper including the cadmium content in coals from the Qiangtang Basin, which was first reported [20] and the elevated concentrations of cadmium in coals from some coalfields, such as the Heshan Coalfield in Guanxi [21–23], the Moxinpo Coalfield in Chongqing [11], etc. The content of cadmium in Chinese coals are presented in Table 2. The arithmetic mean of cadmium in Chinese coals is 0.43 μg/g.

There are six major coal-forming periods in China: The 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) and five coal-distribution areas: The Northeastern area (J_3–K_1 and E–N coals); the Northwestern area ($J_{1–2}$ coals); the Northern area (dominated by C_2–P_1 coals); the Tibet–Western Yunnan area (E–N and T_3 coals), and the Southern area (P_2, T_3 and C_1 coals) [7,17,24]. The coal reserves (according to the Third National Prediction of Coal Resources in China [24]) are listed in Table 3.

Table 2. Cadmium concentration in coals of China.

Coalfields/Province	Sample Number	Mean [1] (μg/g)	Coal-Distribution Area	Coal-Forming Period	CC [2]	Reference
Kailuan/Hebei	48	0.2	Northern	C_2–P_1	0.8	Tang et al. [25], Zhuang et al. [26]
Xingtai/Heibei	2	0.31	Northern	C_2–P_1	1.2	Ren et al. [17]
Huainan/Anhui	50	0.07	Northern	C_2–P_1	0.3	Tong et al. [27], Tang et al. [15], Liu et al. [28], Chen et al. [29], Lu et al. [30]
Huaibei/Anhui	40	0.23	Northern	C_2–P_1	0.9	Chen et al. [31], Jiang et al. [32]
Weibei/Shaanxi	23	0.11	Northern	C_2–P_1	0.4	Yang et al. [33], Wang et al. [34]
Pingdingshan/Henan	1	0.1	Northern	C_2–P_1	0.4	Ren et al. [17]
Datun/Jiangsu	2	0.37	Northern	C_2–P_1	1.5	Zhou et al. [35]
Xuzhou/Jiangsu	5	0.03	Northern	C_2–P_1	0.1	Tang et al. [15]
Ningwu/Shanxi	285	0.2	Northern	C_2–P_1	0.8	Zhao et al. [36], Ren et al. [17], Song et al. [37], Yang et al. [38]
Xishan/Shanxi	21	0.47	Northern	C_2–P_1	1.9	Sun et al. [39]
Hedong/Shanxi	29	0.08	Northern	C_2–P_1	0.3	Ren et al. [17]
Yangquan/Shanxi	6	0.66	Northern	C_2–P_1	2.6	Ren et al. [17]
Jincheng/Shanxi	9	0.16	Northern	C_2–P_1	0.6	Ren et al. [17]
Fenxi/Shanxi	3	1.2	Northern	C_2–P_1	4.8	Zhang et al. [40]
Hunyuan/Shanxi	1	1.2	Northern	C_2–P_1	4.8	Zhang et al. [40]
Lu'an/Shanxi	1	0.05	Northern	C_2–P_1	0.2	Bai [13]
Parts of mines in Shanxi/Shanxi	78	1.09	Northern	C_2–P_1	4.4	Zhang et al. [41]
Jining/Shandong	59	0.39	Northern	C_2–P_1	1.6	Jiang et al. [42], Wang et al. [43]
Juye/Shandong	13	0.4	Northern	C_2–P_1	1.6	Wang et al. [43]
Yanzhou/Shandong	1	0.26	Northern	C_2–P_1	1.0	Bai [13]
Feicheng and Xinwen/Shandong	7	0.27	Northern	C_2–P_1	1.1	Zeng et al. [44]
Shizuishan/Ningxia	10	0.61	Northern	C_2–P_1	2.4	Song et al. [37], Ren et al. [17]
Shitanjing/Ningxia	14	0.43	Northern	C_2–P_1	1.7	Song et al. [37], Ren et al. [17]
Taiyangcheng/Ningxia	3	0.04	Northern	C_2–P_1	0.2	Ren et al. [17]
Baijigou/Ningxia	2	1.23	Northern	C_2–P_1	4.9	Song et al. [37]
Chenjiashan/Shaanxi	8	1.7	Northern	$J_{1\text{-}2}$	6.8	Yang [45]
Huanglong/Shaanxi	46	0.15	Northern	$J_{1\text{-}2}$	0.6	Mo et al. [46]
Shendong/Shaanxi and Inner Mongolia	730	0.03	Northern	$J_{1\text{-}2}$	0.1	Dou et al. [47], Song et al. [37], Wang [48]
Yima/Henan	3	0.71	Northern	$J_{1\text{-}2}$	2.8	Ren et al. [17]
Datong/Shanxi	39	0.17	Northern	$J_{1\text{-}2}$	0.7	Bai [49], Zhang [41], Ren et al. [17], Song D.Y. [37], Liu [50]
Rujigou/Ningxia	3	1.15	Northern	$J_{1\text{-}2}$	4.6	Song et al. [37], Ren et al. [17]
Ciyaopu/Ningxia	1	0.04	Northern	$J_{1\text{-}2}$	0.2	Ren et al. [17]
Huating/Gansu	1	0.08	Northern	$J_{1\text{-}2}$	0.3	Ren et al. [17]
Fanci and Yuanqu/Shanxi	3	2.2	Northern	E–N	8.8	Zhang et al. [41],
Jungar/Inner Mongolia	122	0.11	Northeastern	C_2–P_1	0.4	Yang [51], Yang et al. [52],Xiao et al. [53], Dai et al. [54–56]
Wuda/Inner Mongolia	3	0.22	Northeastern	C_2–P_1	0.9	Ren et al. [17]
Daqingshan/Inner Mongolia	67	0.29	Northeastern	C_2–P_1	1.2	Zou et al. [57], Dai et al. [58]
Baishan District/Jilin	56	0.18	Northeastern	C_2–P_1	0.7	Wu et al. [59]
Beipiao/Liaoning	29	0.28	Northeastern	$J_{1\text{-}2}$	1.1	Kong et al. [60]
Shengli/Inner Mongolia	43	0.07	Northeastern	J_3–K_1	0.3	Dai et al. [61,62]
Baiyinhua/Inner Mongolia	51	0.13	Northeastern	J_3–K_1	0.5	Zhang [63]
Huolinhe/Inner Mongolia	4	0.09	Northeastern	J_3–K_1	0.4	Ren et al. [17], Gao et al. [64]
Dayan/Inner Mongolia	3	0.08	Northeastern	J_3–K_1	0.3	Ren et al. [17]

Table 2. *Cont.*

Coalfields/Province	Sample Number	Mean [1] (μg/g)	Coal-Distribution Area	Coal-Forming Period	CC [2]	Reference
Yimin/Inner Mongolia	8	0.12	Northeastern	J_3–K_1	0.5	Li et al. [65]
Fuxin/Liaoning	3	0.12	Northeastern	J_3–K_1	0.5	Ren et al. [17]
Tiefa/Liaoning	4	0.12	Northeastern	J_3–K_1	0.5	Ren et al. [17]
Hegang/Heilongjiang	3	0.08	Northeastern	J_3–K_1	0.3	Ren et al. [17]
Jixi/Heilongjiang	3	0.127	Northeastern	J_3–K_1	0.5	Ren et al. [17]
Shuangyashan/Heilongjiang	3	0.11	Northeastern	J_3–K_1	0.4	Ren et al. [17]
Qitaihe/Heilongjiang	3	0.27	Northeastern	J_3–K_1	1.1	Ren et al. [17]
Shenbei/Liaoning	2	0.07	Northeastern	E–N	0.3	Ren et al. [17]
Songshao/Yunnan	12	0.36	Southern	C_2–P_1	1.4	Wang [66]
Changguang/Zhejiang	2	0.51	Southern	P_2	2.0	Ren et al. [17]
Shaoguan/Guangdong	1	0.15	Southern	P_2	0.6	Ren et al. [17]
Heshan/Guangxi	51	0.64	Southern	P_2	2.6	Shao et al. [22], Dai et al. [21]
Fusui/Guangxi	19	0.84	Southern	P_2	3.4	Dai et al. [67]
Yishan/Guangxi	22	1.55	Southern	P_2	6.2	Dai et al. [68]
Daye/Hubei	2	0.36	Southern	P_2	1.4	Ren et al. [17]
Yong'an/Fujian	5	0.19	Southern	P_2	0.8	Ren et al. [17]
Chenxi/Hunan	15	5.01	Southern	P_2	20.0	Li et al. [10]
Meitian/Hunan	10	3.8	Southern	P_2	15.2	Tang et al. [15]
Doulishan/Hunan	1	0.38	Southern	P_2	1.5	Ren et al. [17]
Northeastern Jiangxi	44	0.7	Southern	P_2	2.8	Zhuang et al. [69]
Feiling/Jiangxi	2	0.05	Southern	P_2	0.2	Ren et al. [17]
Yinggangling/Jiangxi	1	0.04	Southern	P_2	0.2	Ren et al. [17]
Liuzhi/Guizhou	11	0.42	Southern	P_2	1.7	Zhuang et al. [70], Ren et al. [17]
Shuicheng/Guizhou	58	0.42	Southern	P_2	1.7	Zhuang et al. [70], Ren et al. [17], Feng [71], Zeng et al. [72]
Liupanshui/Guizhou	14	0.12	Southern	P_2	0.5	Qin et al. [73], Feng [71]
Zhijin/Guizhou	24	0.52	Southern	P_2	2.1	Dai, S. et al. [74]
Nayong/Guizhou	6	0.17	Southern	P_2	0.7	Dai, S. et al. [74]
Bijie/Guizhou	3	0.07	Southern	P_2	0.3	Dai, S. et al. [74]
Southwest Guizhou	64	2.46	Southern	P_2	9.8	Song et al. [75], Wei et al. [76], Zhang et al. [77]
Dahebian/Guizhou	12	0.15	Southern	P_2	0.6	Song et al. [78]
Pu'an/Guizhou	9	0.44	Southern	P_2	1.8	Dai et al. [74], Yang [79]
Qinglong/Guizhou	4	0.1	Southern	P_2	0.4	Dai et al. [74]
Zhuzang/Guizhou	2	0.05	Southern	P_2	0.2	Dai et al. [74]
Panjiang/Guizhou	3	0.13	Southern	P_2	0.5	Dai et al. [74]
Dafang/Guizhou	74	0.39	Southern	P_2	1.6	Dai et al. [74,80]
Xingren/Guizhou	6	0.3	Southern	P_2	1.2	Dai et al. [74]
Songzao/Chongqing	26	0.34	Southern	P_2	1.4	Zhao et al. [81], Ren et al. [17], Dai et al. [82]
Nantong/Chongqing	24	0.33	Southern	P_2	1.3	Chen et al. [83]
Moxinpo/Chongqing	8	31.19	Southern	P_2	124.8	Dai et al. [11]
Shiping/Sichuan	6	5.91	Southern	P_2	23.6	Luo et al. [84]
Huayingshan/Sichuan	20	0.25	Southern	P_2	1.0	Zhuang et al. [85]
Xinde/Yunnan	7	0.47	Southern	P_2	1.9	Dai et al. [86]
Taoshuping/Yunnan	17	0.21	Southern	P_2	0.8	Wang et al. [87]
Yantang/Yunnan	24	0.26	Southern	P_2	1.0	Shao et al. [88]

Table 2. *Cont.*

Coalfields/Province	Sample Number	Mean [1] (μg/g)	Coal-Distribution Area	Coal-Forming Period	CC [2]	Reference
Yanshan/Yunnan	7	2.07	Southern	P_2	8.3	Dai et al. [89]
Laochang/Yunnan	42	0.59	Southern	P_2	2.4	Tang et al. [15]
Xuanwei/Yunnan	6	1.24	Southern	P_2	5.0	Dai et al. [90]
Guxu/Yunnan	11	0.56	Southern	P_2	2.2	Dai et al. [91]
Zhenfeng/Guizhou	5	0.26	Southern	T_3	1.0	Zhang et al. [92], Tao et al. [93]
Changhe/Chongqing	16	0.22	Southern	T_3	0.9	Wang et al. [94,95]
Lewei/Sichuan	2	0.1	Southern	T_3	0.4	Ren et al. [17]
Dayi/Sichuan	1	0.2	Southern	T_3	0.8	Ren et al. [17]
Pingxiang/Jiangxi	5	0.48	Southern	T_3	1.9	Ren et al. [17]
Kebao/Yunnan	1	0.4	Southern	E–N	1.6	Ren et al. [17]
Chuxiong/Yunnan	3	0.11	Southern	E–N	0.4	Ren et al. [17]
Huaning/Yunnan	1	0.12	Southern	E–N	0.5	Ren et al. [17]
Kubai/Xinjiang	11	0	Northwestern	$J_{1\text{-}2}$	0.0	Wang et al. [96]
Yili/Xinjiang	77	0.07	Northwestern	$J_{1\text{-}2}$	0.3	Zhao et al. [97], Dai et al. [98]
Juggar/Xinjiang	96	0	Northwestern	$J_{1\text{-}2}$	0.0	Li et al. [99]
Yining/Xinjiang	16	0.04	Northwestern	$J_{1\text{-}2}$	1	Jiang et al. [100]
Muli/Qinghai	18	0.12	Northwestern	$J_{1\text{-}2}$	0.5	Dai et al. [101]
Yuka/Qinghai	1	0.04	Northwestern	$J_{1\text{-}2}$	0.2	Ren et al. [17]
Mole/Qinghai	1	0.01	Northwestern	$J_{1\text{-}2}$	0.0	Ren et al. [17]
Jiangcang/Qinghai	1	0.02	Northwestern	$J_{1\text{-}2}$	0.1	Ren et al. [17]
Datong/Qinghai	1	0.03	Northwestern	$J_{1\text{-}2}$	0.1	Ren et al. [17]
Tumen/Tibet	32	0.47	Tibet-Western Yunnan	T_3	1.9	Fu et al. [20]
Wuruoshan/Tibet	12	0.2	Tibet-Western Yunnan	T_3	0.8	Fu et al. [20]
Hongshuihe/Tibet	6	0.21	Tibet-Western Yunnan	T_3	0.8	Fu et al. [20]
Huaping/Yunnan	1	0.21	Tibet-Western Yunnan	T_3	0.8	Ren et al. [17]
Chuanxi/Sichuan	3	0.37	Tibet-Western Yunnan	E-N	1.5	Ren et al. [17]
Changning/Yunnan	3	0.2	Tibet-Western Yunnan	E-N	0.8	Ren et al. [17]
Lanping/Yunnan	3	0.34	Tibet-Western Yunnan	E-N	1.4	Ren et al. [17]
Lincang/Yunnan	54	0.98	Tibet-Western Yunnan	E-N	3.9	Dai et al. [102]
China	2999	0.43				

[1] Arithmetic average of cadmium in coal; [2] Concentration coefficient, for details see Section 3.1.

Given the uneven coal distribution either in space or in geologic ages, the relative proportion of coal reserves of coal-distribution areas in the corresponding coal-forming period were taken into consideration as weighting factors [7,17]. Some coal samples were lacking in this study, so these related parts were not taken into consideration. Then, the weighted average value of cadmium in coal was estimated as 0.28 μg/g (Table 4), a little higher than the average data given by Ren et al. [17] and Dai et al. [7]. Due to the small coal reserves of these coal-distribution areas, particularly in those coal-forming periods, the cadmium concentration in these samples may have had little impact on the final average. Nevertheless, when those samples were inferred, the weighted mean value of cadmium in Chinese coals may have been a little higher than 0.28 μg/g. However, the value we calculated was still of great significance in understanding the abundance of cadmium in Chinese coals.

Table 3. Coal reserves of China among different coal-distribution areas in different coal-forming periods (unit: 10^9 t).

Coal-Distribution Area	C_2–P_1	P_2	T_3	$J_{1–2}$	J_3–K_1	E–N	Total
Northeastern area	14.64	-	-	24.81	1226.34	45.9	1311.69
Northern area	3829.18	2.12	8.51	2793.94	8.32	14.08	6656.16
Southern area	13.87	761.89	35.92	1.55	-	165.16	978.4
Northwestern area	12.86	0.88	0.14	1207.61	2.08	-	1223.57
Tibet–Western Yunnan area	0.63	0.03	0.19	-	0.04	5.74	6.63
Total	3871.16	765.92	44.77	4027.9	1236.79	230.89	10,176.45

Table 4. The weighted averaging content in Chinese coals (unit: μg/g).

Coal-Bearing Region	Coal-Forming Period	Sample Number	Mean [1]	Coal Reserve Percentage (%)	Weighted Mean Value
Northern area	C_2–P_1	713	0.33	37.6279	0.1245
	P_2	0	-	0.0208	-
	T_3	0	-	0.0836	-
	J_{1-2}	831	0.07	27.4550	0.0181
	J_3–K_1	0	-	0.0818	-
	E–N	3	2.20	0.1384	0.0030
Northeastern area	C_2–P_1	248	0.18	0.1439	0.0003
	J_{1-2}	29	0.28	0.2438	0.0007
	J_3–K_1	128	0.11	12.0508	0.0130
	E–N	2	0.07	0.4510	0.0003
Southern area	C_2–P_1	12	0.36	0.1363	0.0005
	P_2	663	1.27	7.4868	0.0951
	T_3	29	0.26	0.3530	0.0009
	J_{1-2}	0	-	0.0152	-
	E–N	59	0.91	1.6230	0.0148
Northwestern area	C_2–P_1	0	-	0.1264	-
	P_2	0	-	0.0086	-
	T_3	0	-	0.0014	-
	J_{1-2}	222	0.04	11.8667	0.0044
	J_3–K_1	0	-	0.0204	-
Tibet-Western Yunnan area	C_2–P_1	0	-	0.0062	-
	P_2	0	-	0.0003	-
	T_3	51	0.37	0.0019	0.000007
	J_3–K_1	0	-	0.0004	-
	E–N	9	0.30	0.0564	0.00017
China	C_2–N	2999	0.43	1	0.28

[1]: Arithmetic mean of cadmium.

2.2. Comparison with the Cadmium Abundance in the World's Coals

The concentration of cadmium in Chinese coals had a wide range from 0–158 μg/g, but 83% of the samples had a cadmium concentration between 0.0 and 0.5 μg/g. The arithmetic average of the cadmium concentration in coals of China (0.43 μg/g) was slightly lower than that of the American coals calculated by Finkelman (0.47 μg/g) [103], while the resource weighted average cadmium concentration (0.28 μg/g) was much higher than the geometric mean of American coals estimated by Finkelman (0.02 μg/g) [103]. It was also higher than the world average reported by Ketris and Yudovich (0.22 μg/g, 2009) [104]. The main range of Chinese coal was lower than the global range reported by Swaine (0.1–3 μg/g, 1990) [105].

3. Distribution Characteristics of Cadmium in Chinese Coals

Trace elements are present in coals at different concentrations, depending on the various processes by which they have entered the coal at the different stages of coalification [106]. Different geological factors control the migration and enrichment of associated elements in coals [107]. Consequently, there are some obvious differences in the concentrations of associated trace elements in coals from different coal forming periods and coal-distribution areas [16,107]. According to the samples collected, the distribution characteristics of cadmium in Chinese coals are discussed below.

3.1. Distribution Characteristics of Cadmium in Chinese Coals in Different Areas

Based on the collected data of cadmium content in Chinese coals, the arithmetic means of cadmium in coals of different provinces in China are presented in Figure 1 and Table 5. The concentration coefficient (CC, the element concentration in the investigated coals versus the referenced coals: abnormal enrichment (CC > 100), significant enrichment (100 > CC > 10), enrichment (10 > CC > 5), slight enrichment (5 > CC > 2), and depletion (0.5 > CC)), which represents the enrichment level of trace elements in coal, was used to show the enrichment level of cadmium in the coals of China. To avoid the potential error caused by those lacking samples and unavailable first-hand data in this paper, the widely used value of Dai et al. [7] is used as a background value to compare with these investigated coals. The variation characteristics of cadmium in coals of different provinces in China are shown in Figures 1 and 2.

Cadmium in coal is highly enriched in Hunan and Chongqing. It is enriched in Sichuan. Cadmium in coal is slightly enriched in Henan, Ningxia, Yunnan, Zhejiang, Guangxi, Jiangxi and Guizhou. However, cadmium in coal is depleted in Xinjiang, Gansu and Qinghai. The concentrations of cadmium in coals of other provinces (except Beijing, Shanghai, Tianjin, Hainan and Taiwan) are normal (Figures 1 and 2).

The content of cadmium in Chinese coals among the different coal-distribution areas are demonstrated in Table 6. The Tibet-Western Yunnan and the Southern coal-distribution areas both had elevated cadmium concentrations in coals. The Southern area had the highest value, with a particularly high arithmetic average reaching to 1.21 μg/g, nearly five times as high as the value reported by Dai et al. [7]. The coalfields (mines) with abnormally high concentrations of cadmium in coal, such as the Moxinpo Coalfield in Chongqing (31.19 μg/g) [11], the Chenxi Mine in Hunan (5.01 μg/g) [10], the Shiping Mine in Sichuan (5.91 μg/g) [84], are located in this coal-distribution area. In the other coal-distribution areas, cadmium concentrations were all lower than that estimated by Dai et al. [7]. The Northwest area has the lowest concentration of cadmium in coal, with a value of 0.04 μg/g, much lower than Dai et al. [7].

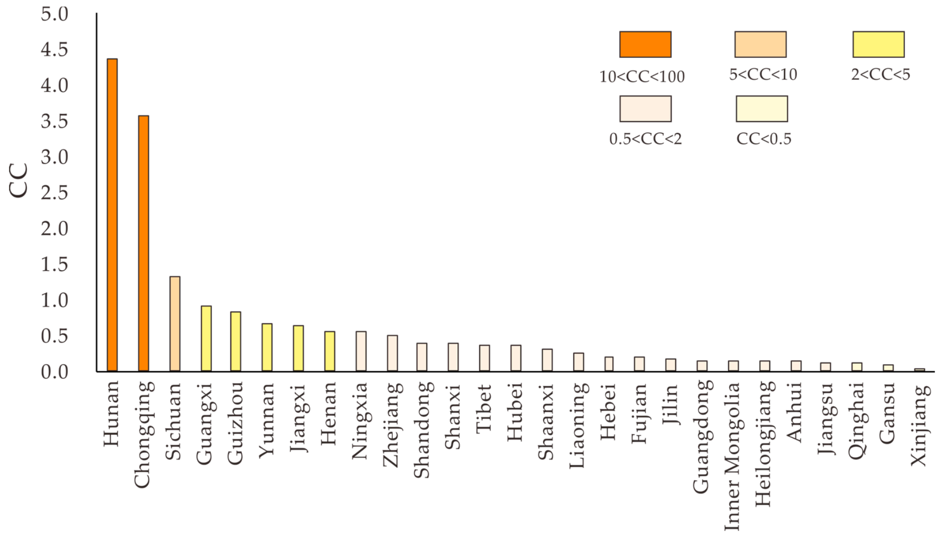

Figure 1. Arithmetic mean values of cadmium in coals from different provinces of China (CC: concentration coefficient, cadmium value from Dai et al. (0.25 μg/g) [7] was used to determine CCs; there were no data in Taiwan, Hainan, Beijing, Tianjin, and Shanghai).

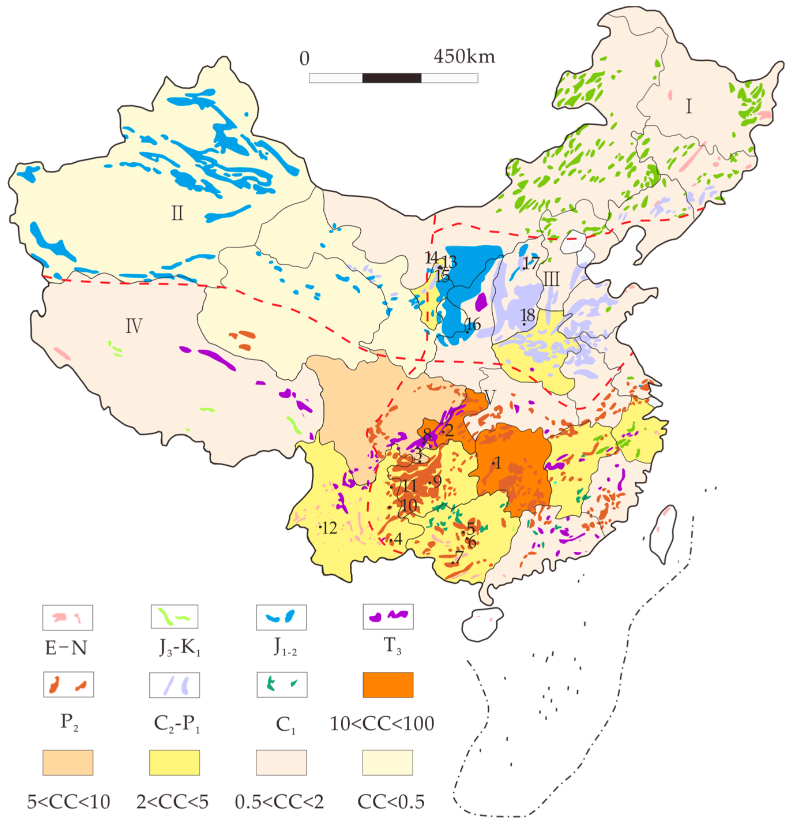

Figure 2. Distribution of cadmium in Chinese coals, modified from Dai et al. [7]. I. Northeastern area; II. Northwestern area; III. Northern area; IV. Tibet–Western Yunnan area; V. Southern area; CC: concentration coefficient, cadmium value from Dai et al. (0.25 μg/g) [7] was used to determine CCs; 1. Chenxi; 2. Moxinpo; 3. Shiping; 4. Yanshan; 5. Yishan; 6. Heshan; 7. Fusui; 8. Guxu; 9. Zhijin; 10. Laochang; 11. Xuanwei; 12. Lincang; 13. Shizuishan; 14. Baijigou; 15. Rujigou; 16. Chenjiashan; 17. Fanci; and 18. Yuanqu. Reproduced with permission from Dai et al; Published by Elsevier, 2012.

Table 5. Cadmium abundance in different provinces in China.

Administrative Division	Coal Reserve/10^9 t *	Sample Number	Mean (μg/g)	CC
Hebei	185.6817	50	0.20	0.8
Anhui	273.5978	90	0.14	0.6
Shaanxi	1554.5631	77	0.30	1.2
Henan	237.9768	4	0.56	2.2
Jiangsu	37.0578	7	0.13	0.5
Shanxi	2500.9125	478	0.38	1.5
Shandong	266.4097	80	0.38	1.5
Ningxia	309.3002	33	0.55	2.2
Gansu	93.0968	1	0.08	0.3
Inner Mongolia	2226.1413	301	0.15	0.6
Jilin	23.0944	56	0.18	0.7
Liaoning	70.6104	38	0.24	1.0
Heilongjiang	200.7548	12	0.15	0.6
Yunnan	240.9296	186	0.67	2.6
Zhejiang	0.0559	2	0.51	2.0
Guangdong	5.7982	1	0.15	0.6
Guangxi	21.841	92	0.90	3.6
Hubei	5.0041	2	0.36	1.4
Fujian	10.6055	5	0.19	0.8
Hunan	33.0624	26	4.37	17.5
Jiangxi	14.0631	52	0.64	2.6
Guizhou	508.0349	295	0.81	3.3
Chongqing [1]	20.45	80	3.65	14.2
Sichuan [2]	117.7701	32	1.31	5.5
Xinjiang	1136.2286	200	0.03	0.1
Qinghai	42.3046	22	0.10	0.4
Tibet	0.9273	50	0.37	1.5

* According to the third coal resource prospecting by the China Coal Geology Bureau [24]; [1] According to Tang et al. [108]; [2]: according to Mao et al. [24] and Tang et al. [108].

Table 6. The arithmetic average of cadmium in different coal-bearing regions.

Coal-Distribution Area	Sample Number	Mean (μg/g)	Coal Reserve Percentage (%)
Northeastern area	407	0.16	12.8
Northern area	1547	0.19	65.4
Northwest area	222	0.04	12
Southern area	709	1.21	9.6
Tibet–Western Yunnan area	114	0.65	0.06

3.2. Distribution Characteristics of Cadmium in Chinese Coals in Different Coal-Forming Periods

Table 7 shows the content of cadmium among the main six coal-forming periods in China. The cadmium content varied considerably among the different coal-forming periods. The late Permian coal-forming period had an abnormal enrichment of cadmium, with a CC as high as 5. The Early and Middle Jurassic and Late Jurassic and Early Cretaceous coal-forming period both had relatively low concentrations of cadmium in coal.

Table 7. The arithmetic average of cadmium in different coal-forming periods.

Coal-Forming Period	Sample Number	Mean (μg/g)	Coal Reserve Percentage (%)
C_2–P_1	973	0.29	38.0
P_2	663	1.27	7.5
J_{1-2}	1082	0.07	12.1
J_3–K_1	128	0.11	39.6
T_3	80	0.33	0.4
E–N	73	0.87	2.3

4. Modes of Occurrence of Cadmium in Chinese Coals

Since coal combustion may be an important source of atmospheric emissions of environmentally relevant trace elements, it is important to know what the levels and modes of occurrence of such elements are in coal [22,85]. Trace elements occur in coals associated with the organic matter and the mineral matter [109]. In general, trace elements are associated with silicate minerals (especially clays), carbonate minerals, sulfide minerals, oxides, and phosphates [109].

Sphalerite is considered to be the predominant carrier of cadmium in coal [9,110]. Furthermore, cadmium is also found in organic components [111], pyrite [105], clays, and carbonates [112]. Quantification of the modes of occurrence of cadmium in low-rank coal was concluded by Finkelman et al. [113]. In low-rank coals, 80% of the cadmium was associated with monosulfides (primarily sphalerite), 10% with pyrite, and 10% with the silicates [113].

Many studies on the modes of occurrence of cadmium in Chinese coals have been conducted [10,11,22,36,47,48,62,70,75,79,102,114–118]. Sulfides and silicates are primarily the hosts of cadmium in coals of China. Additionally, cadmium in Chinese coals is also associated with organic matter, carbonates, and other minerals.

4.1. Sulfides Minerals Association

Sulfide minerals are the most common inorganic sulfur form in coal [119]. Cadmium in coal commonly shows sulfide affinity in Chinese coals [10,11,22,36,47,48,70,79,102]. This may be one reason why high sulfur content coal always has a high content of cadmium. As the most popular cadmium carrier, sphalerite is also found in Chinese coals. Sphalerite has been observed in the secretinite lumens of K_1 Coal from the Moxinpo Coalfield [11]. These sphalerites may illustrate the unusually enriched cadmium (CC > 200) in K_1 Coal. In addition, Dai [102] indicated that cadmium in coals from the Dazhai Mine in Yunnan may occur in minor sulfides such as sphalerite, and the correlation coefficient between zinc and cadmium was as high as 0.95.

Besides sphalerite, as the predominant sulfide mineral in coal [119], direct evidence on pyrite associated cadmium have also been collected. Cadmium has been detected in pyrite in coals from Pingshuo Mine [36] in Shanxi and Pu'an Mines [79] in Guizhou by SEM-EDX (Scanning electron microscope with energy dispersive x-ray spectroscopy). By graphite furnace atomic absorption spectrometry (GF-AAS), Zhang also detected cadmium in pyrites in coals from Southwestern Guizhou, where the content was as high as 0.38 μg/g [92].

4.2. Silicate Minerals Association

Silicate minerals are the other main cadmium carrier in Chinese coals. Clays are the primary type of silicate minerals in coal. Cadmium in the No.6 coals from the Donglin Coal Mine in Chongqing was significantly correlated to Al_2O_3 [83], with a Pearson's correlation coefficient of 0.83, suggesting that cadmium in the No. 6 coals was possibly in combination with the clays. Cadmium in the coals of Wulantuga in Inner Mongolia had a correlation coefficient with aluminosilicate higher than 0.7, indicating an aluminosilicate affinity [62]. Furthermore, cadmium was detected in clays in No. 9 Coal in the Pingshuo Mine by SEM-EDX [36].

4.3. Organic Matter Association

It is rare to find cadmium associated with organic matter. Elements with positive correlations with the ash yields indicate an inorganic association suggesting that the elements are combined in minerals in the coal [22]. In most cases, cadmium in coal positively correlated with the ash yield. However, organic associated cadmium has been found in low rank coals in China. By sequential extract, Zhao et al. [116] suggested that cadmium showed some organic affinity in low rank coals from the Yan'an Coal Seam and Shenbei Coalfield. Moreover, Xu [117] found that organic-bound cadmium accounted for 21% in lignite coal from the Shenbei Coalfield by sequential extract. Perhaps organic-bound cadmium primarily occurs in low rank coals.

4.4. Other Minerals Association

Besides sulfides, silicates, and organic matter, cadmium may also occur in carbonates [118]. Additionally, Ding et al. [114] discovered a new mineral mainly constituted by cadmium and chlorine in high-arsenic coal in Southwestern Guizhou.

Cadmium usually shows a positive correlation with sulfur content in coal, but there is no necessary connection. Besides sulfides, it can also combine with silicates and other minerals. An organically bound form also exists in low rank coals. The modes of occurrence of cadmium in Chinese coals are quite complex.

5. Genetic Factors of Cadmium Enrichment in Chinese Coals

In different coal basins and coal-forming periods, there are generally one or a number of geological factors that may influence the enrichment of trace elements in coals [82]. Dai et al. found five genetic enrichment types of trace elements: Source-rock-controlled; marine-environment-controlled; hydrothermal-fluid-controlled; groundwater-controlled; and volcanic-ash-controlled types [7].

Table 8 and Figure 2 list these coalfields with an appreciable content of cadmium (CC > 2). From the table, we can conclude that most of the coals showing a cadmium abnormality had high sulfur content. This was also common within the different samples collected from the same coal mines, e.g., the arithmetic of cadmium in the M7 Coal with a $S_{t,d}$ value of 2.77% from the Yanshan Coalfield was 0.47 μg/g, while the value was 2.07 μg/g in the M9 coals with a $S_{t,d}$ value of 10.65% [89]. Cadmium most likely accumulates in the coal with high sulfur content.

Table 8. Coalfields with appreciable content of cadmium (CC > 2).

Coalfields/Province	$S_{t,d}$ [1]	CC	Coal-Distribution Area	Period	Reference
Chenxi/Hunan	9.48	20.0	Southern	P_2	Li et al. [10]
Southwest Guizhou	6.98	9.8	Southern	P_2	Song et al. [75]
Moxinpo/Chongqing	2.89	124.8	Southern	P_2	Dai et al. [11]
Shiping/Sichuan	2.79	23.6	Southern	P_2	Luo et al. [84]
Yanshan/Yunnan	10.65	8.3	Southern	P_2	Dai et al. [89]
Yishan/Guangxi	8.74	6.2	Southern	P_2	Dai et al. [68]
Heshan/Guangxi	7.9	2.6	Southern	P_2	Dai et al. [21]
Fusui/Guangxi	6.56	3.4	Southern	P_2	Dai et al. [67]
Guxu/Yunnan	2.73	2.2	Southern	P_2	Dai et al. [91]
Zhijin/Guizhou	1.15	2.1	Southern	P_2	Dai et al. [74,120]
Laochang/Yunnan	>2.5	2.4	Southern	P_2	Tang et al. [15], Yang [121]
Xuanwei/Yunnan	0.18	5.0	Southern	P_2	Dai et al. [90]
Lincang/Yunnan	1.78	3.9	Tibet–Western Yunnan	P_2	Dai et al. [102]
Shizuishan/Ningxia	3.13	2.4	Northern	C_2–P_1	Song et al. [37]
Baijigou/Ningxia	0.14	4.9	Northern	C_2–P_1	Song et al. [37]
Rujigou/Ningxia	0.08	4.6	Northern	$J_{1–2}$	Song et al. [37]
Chenjiashan/Shaanxi	-	6.8	Northern	$J_{1–2}$	Yang et al. [45]
Fanci and Yuanqu/Shanxi	-	8.8	Northern	E–N	Zhang et al. [41]

[1] Total sulfur on dry basis.

However, there are still some exceptions. By comparing high-sulfur coal samples ($S_{t,d}$: 2.15%–4.20%) and low-sulfur coal samples ($S_{t,d}$: 0.50%–1.22%) in Jining in Shandong Province, the values of cadmium in both samples had no distinct difference and were high in both coal samples. Zhuang et al. [69] compared the Late Permian high-sulfur coal with the Late Triassic low-sulfur coal from Northeast Jiangxi Province, and the late had more cadmium. There are many factors that may influence the cadmium enrichment in coal.

5.1. Marine-Environment-Controlled Cadmium Enrichment

High-sulfur coals are usually deposited in a seawater-influenced environment [22,119]. According to the distribution characteristic of cadmium in Chinese coals in space and in geological age, cadmium was mostly enriched in the Southern area during the Late Permian. (Tables 2 and 5–7). The coals with a high cadmium content (CC > 2) were all medium–high-sulfur coals and high-sulfur coals (according to Chinese National Standard (GB/T 15224.2–2010) [122]: <0.5% for ultra-low-sulfur coal, 0.51% to 0.9% for low-sulfur coal, 0.91% to 1.50% for medium sulfur coal, 1.51%–3% for medium-high-sulfur coal, and >3% for high-sulfur coal) except for the coals from Xuanwei (Figure 2). Although it is relatively unusual for coal to be preserved within marine carbonate successions [22], these coals were all preserved within marine carbonate successions in Southern China [22,68]. This may be a reason for the enrichment of cadmium in coals from the Southern area in the Late Permian.

5.2. Hydrothermal-Fluid-Controlled Cadmium Enrichment

The hydrothermal-fluid-controlled type includes magmatic-, low temperature-hydrothermal-fluid-, and submarine-exhalation controlled subtypes [7]. Dai et al. [89] indicated that the strong enrichment of sulfur at the M9 Coal from the Yanshan Coalfield was attributed to the influence of submarine exhalation rather than the marine environment based on the discovery of albite, plagioclase, and dawsonite, which were formed by hydrothermal fluids probably associated with submarine exhalation which invaded along with seawater into the anoxic peat swamp. The cadmium concentration in the coal from the Meitian Coal Mine in Hunan decreased with the increasing distance from the Qitianling intrusive rock [123].

5.3. Source-Rock-Controlled Cadmium Enrichment

In some small fault-controlled basins or coalfields, the sediment-source region can be a dominant factor in trace-element enrichment in coal [7,16]. Elements with high concentrations in the source rocks are commonly enriched in these coals [16].

The coal beds from Fanci County and Yuanqu County in Shanxi are deposited in small intermountain fault basins [41]. Many trace elements are highly enriched and the cadmium concentration was 2.2 μg/g in the coal from this region. Basalt occurs in broad areas of the basins. Zhang suggested that this basalt may be a source of some potential hazardous trace elements including cadmium in the Tertiary brown coals during coal deposition [41].

5.4. Volcanic-Ash-Controlled Cadmium Enrichment

Volcanic ash, which is usually referred to as tonstein, can affect trace element concentrations in coal [124]. Elements may be leached out from the tonstein and are then incorporated into the organic matter. Moreover, volcanic ash located either on the margin of the coal basin or on uplifts within the basin may provide a source of terrigenous material [124]. Influences of volcanic ash on trace elements in coal were mainly reported from Southwestern China [7].

The No. 12 Coal from the Songzao Coalfield had a cadmium content of 0.68 μg/g, while the other coals had an average value of 0.47 μg/g. Compared to the other coals, the No. 12 Coal was deposited directly above the mafic tuff beds [82]. Due to the marine transgression over peat deposits and abundant Fe derived from the underlying mafic tuff bed, the No. 12 Coal has a significantly high content of pyrite (8.1% on average), which caused a high cadmium concentration.

5.5. Groundwater-Controlled Cadmium Enrichment

The groundwater can adsorb these trace elements from the layers directly overlying or underlying the coal seam, carry them into the coal seam, and make some elements accumulate in the coal seam. In addition, the groundwater can also leach some elements to reduce these concentrations in coal [118]. The high cadmium concentration of the No. 9 Coal from the Pingshuo Surface Mine in Shanxi was related to the high concentrations of calcite deposited from the groundwater [118].

6. Conclusions

(1) The arithmetic mean cadmium content in coals of China was 0.43 μg/g. Take the coal reserves into consideration, the weighted average value of cadmium in coal was estimated as 0.28 μg/g, a little higher than the world average.

(2) Cadmium was highly enriched in the coals formed during the Late Permian in the Southern area. It was highly enriched in Hunan and Chongqing, and enriched in Sichuan. Cadmium in coals formed in the Early and Middle Jurassic from the Northwestern area was depleted. Xinjiang, Qinghai, and Gansu had the cleanest coals associated with cadmium.

(3) The modes of occurrence of cadmium in Chinese coals are quite complex. Cadmium showed a positive correlation with sulfur content in coal, indicating a sulfide affinity. The sulfides and silicates were the primary hosts of cadmium in the coals of China. Organically associated cadmium was identified in low-rank coals. Furthermore, carbonates and other minerals also contained cadmium in some coalfields.

(4) The marine environment had a huge impact on the cadmium accumulation in Southern area during the Late Permian. In addition, hydrothermal fluids, source rocks and volcanic ash also influenced the content of cadmium in some coalfields in China.

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