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Distribution, modes of occurrence, and main factors influencing lead enrichment in Chinese coals

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Abstract Lead is a potentially harmful element that has caused serious environmental pollution during its mining and use along with serious human health problems. This study assessed lead in Chinese coals based on published literature, with a particular focus on data reported since 2004. The analysis included 9447 individual samples from 103 coalfields or mines in 28 provinces in China. The arithmetic mean content of lead in the studied coals was 15.0 μg/g. Considering the coal reserves, the weighted-average lead concentration in Chinese coals was calculated to be 19.6 μg/g. Lead was significantly enriched in the coals from Henan Province and enriched in the coals from the Tibet Autonomous Region. The coals from Tibet–Western Yunnan and the southern areas of China had elevated lead concentrations. Sulfides are the primary hosts of lead in Chinese coals, although other hosts include silicates, organic matter, carbonates, and other minerals. Source rocks in the sediment-source region and marine environments may be the most significant factors contributing to lead enrichment in Chinese coals. Hydrothermal fluids and peat-forming plants also contribute to lead enrichment in some Chinese coals.

Keywords Lead · Chinese coal · Enrichment · Distribution · Modes of occurrence

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1 Introduction

In 2017, China produced 1747.2 million tons of oil equivalents (mtoe) of coal, which represents a 3.6% increase from 2016 and accounted for 46.4% of the world's total coal production. China consumed 1892.6 mtoe of coal in 2017, which was 0.5% more than in 2016 and accounted for 50.7% of world coal consumption (BP 2018). According to the Statistical Communique on China's 2017 National Economic and Social Development (National Bureau of Statistics 2018), 60.4% of China's energy consumption rely on coal in 2017, and no major change in this percentage is foreseen in the near future. Given the rapid economic development in China, coal will continue to be a major component of China's energy structure. Unfortunately, coal contains many potentially toxic elements such as arsenic, mercury, fluorine, selenium, and lead (Dai and Ren 2007; Finkelman et al. 2018), some in dangerously high concentrations. Therefore, reducing coal consumption to mitigate damage to both human health and the environment is desirable but difficult to achieve.

There are several anthropological sources of lead in the environment, including leaded gasoline, white paint, water pipes and solder, batteries, shotgun pellets, and sulfide mining and weathering. However, lead from these sources is not as widespread as lead emitted from coal combustion. While lead and other trace elements generally have low concentrations in coal, they are rich in coal fly ash by approximately a factor of 10 (Dai et al. 2010b). Widory et al. (2010) found that metal refining plants are the major source of atmospheric lead in Beijing followed by thermal power stations and other coal combustion processes. Studies in China have shown that high lead concentrations may be found in fine particulate matter with diameters less than 2.5 μm ($PM_{2.5}$) and inhalable particles with diameters less than 10 μm (PM_{10}); both $PM_{2.5}$ and PM_{10} can penetrate deep into the lungs of humans and animals (Li and Lin 2006; Tang and Huang 2004; Yang et al. 1987). Approximately 40% of inhaled lead enters the blood circulation system (Shah et al. 2010).

Coal use is a major anthropogenic source of lead in the environment in China and elsewhere. Hu et al. (2011) determined that the primary source of atmospheric lead in China and particularly in southern China is the combustion of lead-containing coal rather than leaded gasoline. Fang et al. (2014, 2015) discovered that lead may be leached from coarse coal refuse under natural weathering conditions, after which it is enriched in local field crops. Leaded paint and leaded gasoline were major sources of lead in the environment until several decades ago, when their use was discouraged, restricted, or banned. Lead pollution in food and drinking water is an important pathway for human lead exposure. The standards for drinking water quality in China (GB 5749–2006) (Ministry of Health of the People's Republic of China 2006) stipulate that the maximum allowable limit of lead in drinking water is 0.01 mg/L.

Lead has been described as a 'multimedia pollutant' due to the numerous and diverse sources and pathways of potential exposure; lead is toxic and accumulates easily in the human body (Bellinger 2008; Richard 2004). The exposure of infants to excess lead can cause mental impairment, diminished IQ, and even blindness. A major source of child lead poisoning, which remains a serious problem in the United States, is the lead that was once used in white paint (Markowitz and Rosner 2000; Pirkle et al. 1998). Montgomery and Mathee (2005) indicated that the weathering, peeling, or chipping of lead-based paint may play an important role in childhood lead exposure in South Africa. Between April and May of 2015, 28 children died as a result of the unregulated, rudimentary processing of lead-rich gold ores in Niger State, Nigeria (Martin 2016). There are many sources of lead poisoning in children; removing lead from paints is only one of the important measures required to reduce exposure (Lin et al. 2009; Turner and Solman 2016). The effects of lead on the peripheral nervous system are more pronounced in adults, whereas the central nervous system is more affected in children (Bellinger 2008; Richard 2004). Moreover, lead may alter gene expression (Rossman 2000). Silbergeld et al. (2000) investigated the oncogenic mechanism of lead and found that lead does not directly change DNA but increases the risk of cancer by reducing the ability of cells to repair their DNA.

In recent years, health studies on lead in coal have focused on the harmful effects of lead exposure, especially in children (Dai et al. 2010a; Song et al. 2012). Therefore, the aggregation of data to determine the physical and chemical characteristics of lead along with the causes of lead enrichment in Chinese coals might be helpful for minimizing the health problems caused by lead in Chinese coal. Based on published data with a focus on studies reported since 2004, this paper provides a comprehensive review of lead in Chinese coals formed during the main coal-forming periods in the primary coal-distribution areas. Based on the aggregated data, we discuss the lead concentration, distribution characteristics, modes of occurrence, and primary factors influencing the enrichment of lead in Chinese coal.

2 Data sources and analysis method

China has not completed a national survey on trace elements in coal. However, a number of published reports contain useful information on the occurrence of lead in Chinese coals. Based on the published literature and with a focus on data reported since 2004, 9447 individual samples from 103 coalfields or mines in 28 provinces were evaluated in this study.

Six major coal-forming periods have occurred 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). The five coal-distribution areas in China are defined as (Fig. 1): 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) (Dai et al. 2012). As shown in Table 1, 8422 samples were from J_{1-2} coals, 522 samples were from C_2–P_1 coals, and 350 samples were from P_2 coals. These three coal-forming periods account for 98.4% of the analyzed data. A major objective of this study was to summarize the content and distribution of lead in Chinese coals from these three main coal-forming periods.

We first verified that the dates of the coal lead data obtained from the literature were within the past few

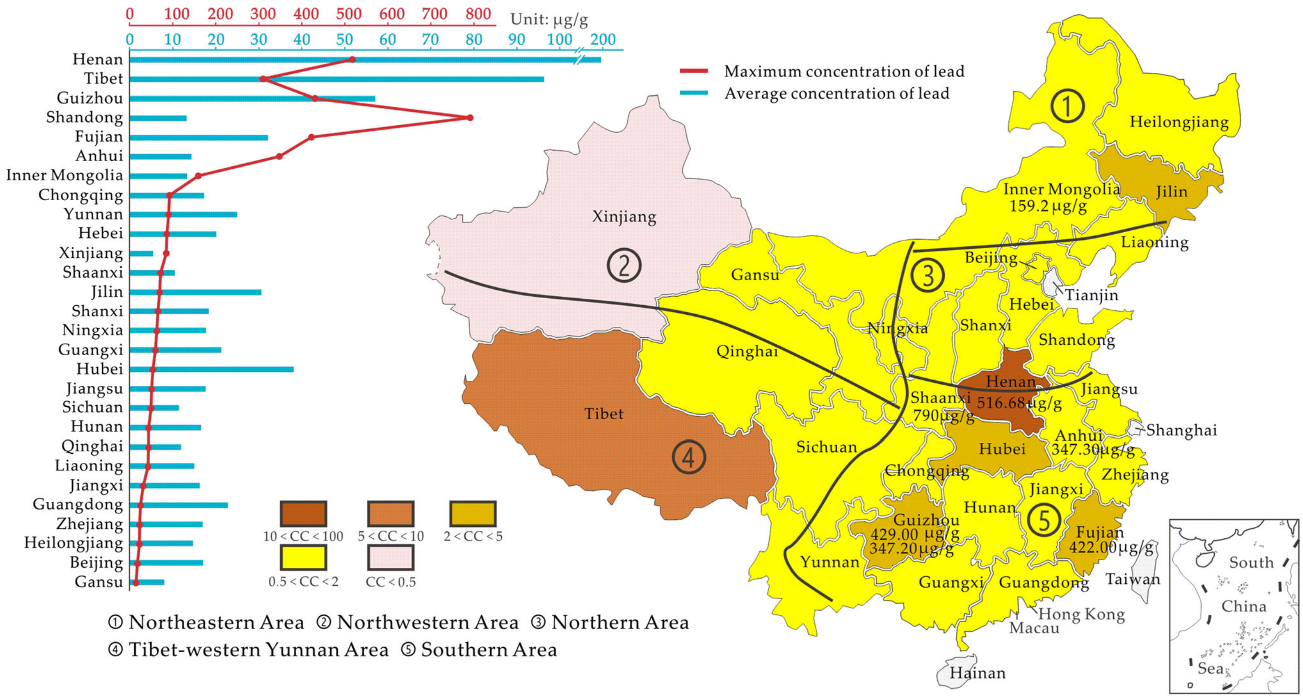

Fig. 1 Distribution of lead in Chinese coals. CC: concentration coefficient

Table 1 Lead concentrations in Chinese coals from the main coal-forming periods

Coal-forming period	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic mean (μg/g)
C_2–P_1	522	0.20	517	30.3
P_2	350	1.6	422	26.0
T_3	44	4.0	72.4	24.5
$J_{1\text{-}2}$	8422	< 0.20	790	12.7
J_3–K_1	62	2.4	310	79.4
E–N	47	2.8	89.6	29.2
Total	9447	< 0.20	790	15.0

decades by tracing the source of the cited data. We then recorded the sampling location, coal-forming period, and coal-forming environment. Finally, the data were grouped by province, coal-distribution area, and coal-forming period for further analysis. The contents of lead in Chinese coals are presented in Table 2. We did not critically evaluate the analytical methodology used in each source. However, we expect that the large number of samples included in this analysis should compensate for a few questionable analyses. The arithmetic mean of lead in the 9447 Chinese coals was 15.0 μg/g. Information on lead in coals throughout the world is also provided for comparison.

3 Contents and modes of occurrence of lead in coals

3.1 World coals

3.1.1 Content of lead in world coals

The contents of lead in coals throughout the world were mostly in the range of 10.0–20.0 μg/g; a few coal samples had lead contents exceeding 100 μg/g, and even fewer reached 1000 μg/g (Table 3). Swaine (1990) reported a range of 2.0–80.0 μg/g for world coals. The Clarke values of lead calculated by Ketris and Yudovich are 6.6 ± 0.4 for world brown coals, 9.0 ± 0.7 for world hard coals, and 7.8 for all coals (Ketris and Yudovich 2009). Based on 7469 coal samples, the arithmetic mean, geometric mean,

Table 2 Lead concentrations in Chinese coals

Coalfield/province	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic mean (μg/g)	Coal distribution area	Coal-forming period	CC[a]	References
Tonghua/Jilin	56	1.6	69.0	30.7	Northern	C_2–P_1	2.0	Wu and Zhou (2004)
Wuda/Inner Mongolia	3	6.9	31.0	19.3	Northern	C_2–P_1	1.0	Dai et al. (2002)
Zhungeer/Inner Mongolia	7	30.5	62.2	38.7	Northern	C_2–P_1	2.7	Yang (2008)
Baotou/Inner Mongolia	33	7.0	77.3	25.6	Northern	C_2–P_1	1.7	Zhou et al. (2013)
Heidaigou/Inner Mongolia	29	9.5	70.0	30.3	Northeastern	C_2–P_1	2.0	Dai et al. (2008)
Kailuan/Hebei	77	7.9	52.7	22.6	Northern	C_2–P_1	1.5	Ren et al. (2006), Zhuang et al. (1999)
Fengfeng/Hebei	5	0.31	21.0	11.3	Northern	C_2–P_1	0.7	Dai (2002), Ren et al. (2006)
Xingtai/Hebei	4	3.4	14.5	11.0	Northern	C_2–P_1	0.7	Ren et al. (2006)
Pingdingshan/Henan	31	10.4	517	199	Northern	C_2–P_1	13.2	Feng et al. (2015), Ren et al. (2006), Tang and Huang (2004)
Xinwen/Shandong	6	5.6	29.7	17.1	Northern	C_2–P_1	1.1	Ren et al. (2006)
Yanzhou/Shandong	5	9.4	21.5	16.1	Northern	C_2–P_1	1.1	Bai (2003), Ren et al. (2006)
Jining/Shandong	38	11.5	36.5	17.5	Northern	C_2–P_1	0.8	Ren et al. (2006)
Taozao/Shandong	8	5.2	26.1	14.4	Northern	C_2–P_1	1.0	Tang et al. (2002)
Huainan/Anhui	5	7.8	26.0	14.1	Northern	C_2–P_1	0.9	Tang et al. (2002)
Huaibei/Anhui	12	5.2	33.1	23.0	Northern	C_2–P_1	1.0	Tang et al. (2002)
Xuzhou/Jiangsu	4	2.3	50.6	17.8	Northern	C_2–P_1	1.2	Ren et al. (2006), Tang et al. (2002)
Hancheng/Shaanxi	23	18.0	42.2	23.0	Northern	C_2–P_1	1.5	Ren et al. (2006)
Tongchuan/Shaanxi	11	16.2	28.7	24.5	Northern	C_2–P_1	1.6	Ren et al. (2006)
Pingshuo/Shanxi	70	0.20	47.4	14.4	Northern	C_2–P_1	1.0	Bai (2003), Ren et al. (2006), Tang et al. (2002)
Hedong/Shanxi	28	2.9	64.7	24.1	Northern	C_2–P_1	1.6	Ren et al. (2006)
Huozhou/Shanxi	10	5.2	28.7	13.6	Northern	C_2–P_1	0.9	Tang et al. (2002)
Xishan/Shanxi	14	19.0	37.0	26.1	Northern	C_2–P_1	1.7	Ren et al. (2006)
Datong/Shanxi	14	12.3	48.1	26.8	Northern	C_2–P_1	1.8	Liu et al. (2013a)
Shizuishan/Ningxia	7	3.8	16.2	8.5	Northern	C_2–P_1	0.6	Dai (2002), Ren et al. (2006)
Shitanjing/Ningxia	13	3.9	39.4	13.4	Northern	C_2–P_1	0.9	Dai (2002); Ren et al. (2006)
Jinzhushan/Hunan	1	43.9	43.9	43.9	Southern	C_2–P_1	2.9	Ren et al. (2006)
Lengshuijiang/Hunan	1	26.5	26.5	26.5	Southern	C_2–P_1	1.8	Ren et al. (2006)
Kaili/Guizhou	7	89.0	429	246	Southern	C_2–P_1	16.3	Wu et al. (2008)
Longtan Formation/Anhui	34	6.1	347	15.7	Southern	P_2	1.0	Qian and Yang (2003)
Changguang/Zhejiang	2	11.6	22.9	17.1	Southern	P_2	1.1	Ren et al. (2006)
Songzao/Chongqing	1	18.1	18.1	18.1	Southern	P_2	1.2	Ren et al. (2006)

Table 2 continued

Coalfield/province	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic mean (μg/g)	Coal distribution area	Coal-forming period	CC[a]	References
Zhongliangshan/Chongqing	1	22.5	22.5	22.5	Southern	P_2	1.5	Ren et al. (2006)
Chuandongqu/Chongqing	2	21.9	92.1	57.0	Southern	P_2	3.8	Ren et al. (2006)
Tianfu/Chongqing	3	16.8	42.5	36.1	Southern	P_2	2.4	Ren et al. (2006), Zhuang et al. (2003)
Donglin/Chongqing	26	1.6	27.1	8.5	Southern	P_2	0.6	Chen et al. (2015)
Moxinpo/Chongqing	4	10.4	25.3	14.3	Southern	P_2	0.9	Dai et al. (2017a)
Junlian/Sichuan	23	2.2	49.7	14.7	Southern	P_2	1.0	Luo (2014), Ren et al. (2006)
Furong/Sichuan	18	1.7	27.8	10.1	Southern	P_2	0.7	Luo (2014), Ren et al. (2006)
Huayingshan/Sichuan	13	5.7	24.5	11.8	Southern	P_2	0.8	Luo (2014)
Guxu/Sichuan	11	1.7	19.3	5.2	Southern	P_2	0.3	Dai et al. (2016)
Panxian/Guizhou	14	7.1	347	45.9	Southern	P_2	3.0	Dai et al. (2005), Guo et al. (1996), Ren et al. (2006)
Liuzhi/Guizhou	11	8.8	32.2	13.6	Southern	P_2	0.9	Guo et al. (1996), Ren et al. (2006), Zhuang et al. (1999)
Zhijin/Guizhou	15	3.8	24.0	13.6	Southern	P_2	0.9	Dai et al. (2005)
Shuicheng/Guizhou	36	6.1	147	20.9	Southern	P_2	1.4	Guo et al. (1996), Ren et al. (2006), Zhuang et al. (1999)
Bijie/Guizhou	33	8.9	184	101	Southern	P_2	6.7	Cheng et al. (2013), Dai et al. (2005)
Xuanwei/Yunnan	15	6.4	36.3	17.9	Southern	P_2	1.2	Shao et al. (2015)
Xinde/Yunnan	7	12.7	26.9	15.5	Southern	P_2	1.0	Dai et al. (2014)
Fengcheng/Jiangxi	2	6.6	11.0	8.7	Southern	P_2	0.6	Ren et al. (2006)
Leping/Jiangxi	4	8.5	18.0	12.4	Southern	P_2	0.8	Lu et al. (1995), Zhuang et al. (1999)
Dazhi/Hubei	2	22.7	53.8	38.3	Southern	P_2	2.5	Lu et al. (1995), Ren et al. (2006)
Xingmei/Guangdong	1	21.5	21.5	21.5	Southern	P_2	1.4	Ren et al. (2006)
Shaoguan/Guangdong	1	24.4	24.4	24.4	Southern	P_2	1.6	Lu et al. (1995)
Heshan/Guangxi	14	4.7	59.0	23.4	Southern	P_2	1.5	Dai et al. (2013a), Lu et al. (1995), Zeng et al. (2005)
Lianshao/Hunan	1	21.8	21.8	21.8	Southern	P_2	1.4	Ren et al. (2006)
Chenxi/Hunan	11	5.2	40.8	12.8	Southern	P_2	0.8	Li et al. (2013)
Yong'an/Fujian	5	5.0	422	32.3	Southern	P_2	2.1	Lu et al. (1995), Ren et al. (2006)
Anjialing/Shanxi	18	4.7	61.3	21.4	Northern	P_2	1.4	Wang et al. (2015)
Helanshan/Ningxia	7	30.5	62.2	38.6	Northern	P_2	2.6	Dai et al. (2006)
Fengfeng-Handan/Hebei	15	8.6	86.6	38.8	Northern	P_2	2.6	Dai and Ren (2007)
Beipiao/Liaoning	3	4.3	42.2	15.9	Northeastern	J_{1-2}	1.1	Kong et al. (2002)
Datong/Shanxi	14	0.90	30.0	11.8	Northern	J_{1-2}	0.8	Ren et al. (2006)
Daanshan/Beijing	1	17.2	17.2	17.2	Northern	J_{1-2}	1.1	Ren et al. (2006)
Rujigou/Ningxia	2	6.4	6.6	6.5	Northern	J_{1-2}	0.4	Ren et al. (2006)
Huating/Gansu	1	2.8	2.8	2.8	Northern	J_{1-2}	0.2	Ren et al. (2006)

Table 2 continued

Coalfield/province	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic mean (μg/g)	Coal distribution area	Coal-forming period	CC[a]	References
Yima/Henan	1	12.9	12.9	12.9	Northern	J_{1-2}	0.9	Lv et al. (2003)
Weixian/Hebei	33	2.7	37.2	8.6	Northern	J_{1-2}	0.6	Chu (2014)
Yaojie/Gansu	1	13.5	13.5	13.5	Northwestern	J_{1-2}	0.9	Ren et al. (2006)
Mole/Qinghai	1	6.4	6.4	6.4	Northwestern	J_{1-2}	0.4	Ren et al. (2006)
Datong/Qinghai	1	22.6	22.6	22.6	Northwestern	J_{1-2}	1.5	Ren et al. (2006)
Xidatan/Qinghai	26	7.7	43.4	14.9	Northwestern	J_{1-2}	1.0	Yi (2016)
Muli/Qinghai	16	0.94	32.6	7.2	Northwestern	J_{1-2}	0.5	Dai et al. (2015a)
Fukang/Xinjiang	1	6.9	6.9	6.9	Northwestern	J_{1-2}	0.5	Ren et al. (2006)
Hami/Xinjiang	2	3.1	9.9	6.5	Northwestern	J_{1-2}	0.4	Ren et al. (2006)
Badaowan/Xinjiang	6	5.2	25.1	12.5	Northwestern	J_{1-2}	0.8	Yang et al. (2005)
Zhundong/Xinjiang	162	< 0.20	25.0	2.3	Northwestern	J_{1-2}	0.2	Zhuang et al. (2013)
Yili/Xinjiang	37	4.3	84.3	20.0	Northwestern	J_{1-2}	1.3	Dai et al. (2015a, b, c)
Yining/Xinjiang	16	0.44	10.8	2.6	Northwestern	J_{1-2}	0.2	Jiang et al. (2015)
Inner Mongolia	7357	< 0.20	159	13.2	Northeastern	J_{1-2}	0.9	Liu et al. (2012)
Tongchuan/Shaanxi	8	1.2	468	122	Northern	J_{1-2}	8.1	Yang et al. (2008)
Shenfu–Dongsheng/Shaanxi	732	0.30	790	8.4	Northern	J_{1-2}	0.6	Ren et al. (2006)
Chuandongqu/Chongqing	1	32.7	32.7	32.7	Southern	J_{1-2}	2.2	Ren et al. (2006)
Huaping/Yunnan	1	19.3	19.3	19.3	Tibet–Western Yunnan	T_3	1.3	Ren et al. (2006)
Yipinglang/Yunnan	1	15.8	15.8	15.8	Tibet–Western Yunnan	T_3	1.0	Ren et al. (2006)
Zixing/Hunan	1	18.1	18.1	18.1	Southern	T_3	1.2	Ren et al. (2006)
Dazhu/Sichuan	1	19.0	19.0	19.0	Southern	T_3	1.3	Zhuang et al. (2003)
Guangwang/Sichuan	3	12.8	27.5	16.3	Southern	T_3	1.1	Ren et al. (2006)
Yongrong/Chongqing	2	38.1	72.4	51.3	Southern	T_3	3.4	Ren et al. (2006)
Jiangbei/Chongqing	1	40.3	40.3	40.3	Southern	T_3	2.7	Ren et al. (2006)
Pingxiang/Jiangxi	8	12.5	31.0	20.4	Southern	T_3	1.3	Ren et al. (2006)
Zhenfeng/Guizhou	4	4.0	31.2	12.5	Southern	T_3	0.8	Tao et al. (2015)
Heshan/Guangxi	12	8.9	39.4	19.2	Southern	T_3	1.3	Shao et al. (2006)
Fuxian/Shaanxi	10	29.0	57.0	37.3	Northern	T_3	2.5	Zhang et al. (2004)
Hegang/Heilongjiang	1	7.1	7.1	7.1	Northeastern	J_3–K_1	0.5	Ren et al. (2006)
Jixi/Heilongjiang	1	22.5	22.5	22.5	Northeastern	J_3–K_1	1.5	Ren et al. (2006)
Huolinhe/Inner Mongolia	2	9.0	9.1	9.1	Northeastern	J_3–K_1	0.6	Bai (2003), Ren et al. (2006)
Yimin/Inner Mongolia	6	2.4	14.3	5.4	Northeastern	J_3–K_1	0.4	Liang et al (2013)

Table 2 continued

Coalfield/ province	Sample number	Min (µg/g)	Max (µg/g)	Arithmetic mean (µg/g)	Coal distribution area	Coal-forming period	CC[a]	References
Tiefa/Liaoning	2	13.3	16.2	14.1	Northeastern	J_3–K_1	0.9	Bai (2003), Ren et al. (2006)
Qiangtang/Tibet	50	11.9	310	96.4	Tibet–Western Yunnan	J_3–K_1	6.4	Fu et al. (2013)
Shulan/Jilin	1	34.5	34.5	34.5	Northeastern	E–N	2.3	Ren et al. (2006)
Huangxian/Shandong	2	17.5	17.9	17.7	Northwestern	E–N	1.2	Ren et al. (2006)
Xiaolongtan/Yunnan	5	2.8	20.9	7.8	Southern	E–N	0.5	Ren et al. (2006), Tang et al. (2002)
Lvhe/Yunnan	2	12.2	39.3	25.7	Tibet–Western Yunnan	E–N	1.7	Ren et al. (2006)
Mengtuo/Yunnan	11	6.9	76.9	34.6	Tibet–Western Yunnan	E–N	2.3	Chen et al. (2016)
Dazhai/Yunnan	26	3.5	89.6	32.1	Tibet–Western Yunnan	E–N	2.1	Dai et al. (2015b)
China	9447	< 0.20	790	15.0				

[a] Concentration coefficient; see Sect. 4.1

and maximum lead contents in United States (U.S.) coals were calculated to be 11.0, 5.0, and ~ 1900 µg/g, respectively (Finkelman 1993). Tang and Huang (2004) calculated arithmetic mean and geometric mean lead contents of 11.1 and 3.5 µg/g, respectively, for world coals. These values are lower than the arithmetic mean of lead in the earth's crust, which was reported as 14.0 µg/g with a range of 12.0–16.0 µg/g (Li 1992).

3.1.2 Modes of occurrence of lead in coals worldwide

Statistical analyses indicated that lead was positively correlated with sulfur and primarily occurred in the form of sulfides (Finkelman 1994), including pyrite (Wang et al. 1997, 2007; Wu and Zhou 2004; Wu et al. 2004) and galena (Ren et al. 2006). The lead content of pyrite in coal was as high as 730 µg/g (White et al. 1989). Compared to bituminous coal, lead was less associated with pyrite in low-rank coal (Finkelman et al. 2018). Galena existed as epigenetic micron-sized particles in coal fractures, pyrite, organic material, and clausthalite (PbSe) (Finkelman 1995). Using a selective leaching protocol on approximately 20 coals, Finkelman et al. (2018) found that 55% of lead in bituminous coals occurred in monosulfides (likely galena) and 35% in pyrite, while only 5%–10% of lead was associated with silicates; in contrast, in low-rank coals, 50% of lead occurred in monosulfides (likely galena), 25% was associated with silicates, and only 10% was in pyrite.

Baruah et al. (2005) reported that lead was enriched in organic macromolecules within high-sulfur coal in northern India. The presence of organic-associated lead in low-rank Malaysian coal was confirmed by electron probe microanalysis (EPMA) (Sia and Abdullah 2012). Carbonate minerals are relatively enriched in middlings, resulting in enrichment in some elements contained in carbonates, including lead (Fu et al. 2018).

In some coal samples, lead is contained in rarer minerals. For example, Finkelman (1981) found that barium could be replaced by lead in barium sulfates, carbonates, phosphates, silicates, and other minerals in coals. Finkelman (1981) noted that substantial amounts of lead in Appalachian Basin coals occur as PbSe, which was confirmed as an important source of lead by X-ray fluorescence, microparticle-induced X-ray emission, energy-dispersive X-ray analysis, and EMPA (Hower and Robertson 2003). Lead and selenium were found to be part of the iron sulfide structure, possibly with selenide substitution in the sulfide structure, as indicated by EPMA (Hower et al. 2008). Li et al. (2001) discovered lead-rich crocoite in New Zealand coals.

Table 3 Contents of lead in world coals

Country	Coalfield		Coal-forming period	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic Mean (μg/g)	References
Canada	Saskatchewan Bienfait mine		E_1	14			6.4	Beaton et al. (1991)
	British Columbian Peace basin		K_1	43	5.9	155	25.1	Van der Flier-Keller et al. (1987)
U.S	Nationwide			7469	1900		11.0	Finkelman (1993)
	Illinois basin		C	175	4.0	237	39.0	Chou (1997)
Iraq	Hemrin South Mountain		N_1	9	11.0	66.7	28.7	Kettanah and Eble (2017)
Australia	New South Wales Gunnedah		P	35	1.1	57.2	9.4	Ward et al. (1999)
England	Main coal fields		C	24	3.2	76.0	16.7	Spears and Zheng (1999)
	South Wales basin		C	26	3.1	76.9	22.5	Gayer et al. (1999)
Turkey	Kozlu basin	Alacaağzı Fm	P_2	154	17.0	61.0	34.0	Karayiğit et al. (2017b)
		Kozlu Fm			2.6	128	34.0	
		Karadon Fm			14.0	315	38.0	
	Nationwide			143[a]	1.0	58.0	9.3	Palmer et al. (2004)
	Çan basin			81	0.70	97.0	9.7	Gürdal (2011)
Czech	North Bohemian basin		N_1	106	3.2	16.0	9.4	Bouška and Pešek (1999)
Greece	Nine coal mines		E_3–Q_1	28	3.7	27.7	13.2	Foscolos et al. (1989)
India	Seven coal mines in the Northeast and northwest parts		E_{2-3}	42	< 1.0	15.0	5.6	Mukherjee et al. (1992)
Poland	Upper Silesian coal basin		E	44	1.3	823	38.8	Smoliński et al. (2014)

[a] Number of mines; Fm: Formation

3.2 Chinese coals

3.2.1 *Content of lead in Chinese coals*

Based on data from 9447 Chinese coal samples, the lowest content of lead was less than 0.20 μg/g, the highest content was 790 μg/g, and the arithmetic mean was 15.0 μg/g (Table 2). The arithmetic mean lead concentration in Chinese coals was higher than in U.S. coals (11.0 μg/g; Table 3) and global coals (3.5 μg/g; Ren et al. 2006). A more practical estimate can be obtained by considering the relative proportion of coal reserves from different regions as weighting factors (Dai et al. 2012; Ren et al. 2006). The weighted-average lead content in Chinese coals was calculated to be 19.6 μg/g (Table 4), higher than those reported by Ren et al. (2004) (15.6 μg/g) and Dai et al. (2012) (15.1 μg/g).

3.2.2 *Modes of occurrence and sources of lead in Chinese coals*

Many papers have been published on lead in Chinese coals (Chen et al. 2009; Cheng et al. 2013; Dai et al. 2002; Finkelman 1994, 1995; Finkelman et al. 2018; Gluskoter et al. 1977; Li et al. 2001; Lv et al. 2003; Yang 2006; Zhao 2015; Zou et al. 2016). These studies generally indicate that sulfides are the primary host of lead in Chinese coals, as in world coals.

Most trace elements in coals are hosted by minerals via isomorphism, adsorption, and interfusion; trace elements are rarely the main component in independent minerals. Fu et al. (2018) found that the proportions of different modes of occurrence of lead in Chinese raw coal decreased in the following order: organic-bound (30%) ≅ silicate-bound (30%) > carbonate-bound (20.4%) > sulfide-bound (19.3%). This distribution differs from our analysis of world coals.

In the Qiangtang Basin of the Tibet Autonomous Region, lead is primarily bound to organic materials, as indicated by the significant negative relationship between lead content and ash yield ($r = -0.73$) along with the positive relationship between lead content and total organic content ($r = 0.63$) (Fu et al. 2013). These relationships are supported by the observations of Zhu (1979) and Gu and Ding (1996), who performed lead ion adsorption experiments in wastewater and found that almost all lead ions were absorbed by peat and lignite. Liang et al. (2013) also found that a weaker correlation between elements and ash yield corresponded to a stronger affinity of organic material for the elements.

Some researchers have reported that lead might exist in aluminosilicates such as clay minerals (Chen et al. 2010; Chu 2014; Querol et al. 1997; Zhao 2015; Zhuang et al. 2013). Sun et al. (2010) and Dai et al. (2018) found that after weathering, lead was enriched in gangue and roofs with high clay contents.

Table 4 Contents of lead in Chinese coals by region

Coal-distribution area	Coal-forming period	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic mean (μg/g)	Coal reserve percentage[a] (%)	Weighted mean value
Northeastern area	C_2–P_1	29	9.5	70.0	30.3	0.1439	0.0436
	$J_{1–2}$	7360	0.20	159	13.2	0.2438	0.0322
	J_3–K_1	12	2.4	22.5	9.0	12.0508	1.0882
	E–N	1	34.5	34.5	34.5	0.4510	0.1556
Northern area	C_2–P_1	484	0.20	517	32.8	37.6279	10.5621
	P_2	40	4.7	86.6	31.0	0.0208	0.0037
	T_3	10	29.0	57.0	37.3	0.0836	0.0312
	$J_{1–2}$	792	0.30	790	9.6	27.4550	2.6439
	J_3–K_1	–	–	–	–	0.0818	–
	E–N	–	–	–	–	0.1384	–
Southern area	C_2–P_1	9	26.5	429	199	0.1363	0.2711
	P_2	310	1.6	422	26.3	7.4868	1.9660
	T_3	32	4.0	72.4	21.0	0.3530	0.0741
	$J_{1–2}$	1	32.7	32.7	32.7	0.0152	0.0050
	E–N	5	2.8	20.9	7.8	1.6230	0.1271
Northwestern area	C_2–P_1	–	–	–	–	0.1264	–
	P_2	–	–	–	–	0.0086	–
	T_3	–	–	–	–	0.0014	–
	$J_{1–2}$	269	0.20	84.3	6.7	11.8667	0.7927
	J_3–K_1	–	–	–	–	0.0204	–
	E–N	2	17.5	17.9	17.7	–	–
Tibet–Western	C_2–P_1	–	–	–	–	0.0062	–
Yunnan area	P_2	–	–	–	–	0.0003	–
	T_3	2	15.8	19.3	17.6	0.0019	0.0003
	J_3–K_1	50	11.9	310	96.3	0.0004	0.0004
	E–N	39	3.5	89.6	32.4	0.0564	0.0183
China	C_2–N	9447			15.0	100.0000	17.82

[a]According to Mao and Xu (1999)

The dominant sources of lead in coal from the Dafang Coalfield were vein ankerite and epigenetic sulfide minerals (Dai et al. 2012). Hower et al. (2003) and Zhao (2015) found that lead in Chinese coals existed in carbonate minerals such as ankerite.

Epigenetic sulfide minerals are the dominant source of lead in Chinese coals (Dai et al. 2012). Lead may occur in pyrite or in trace sulfide minerals (Dai et al. 2008). Lead content and total sulfur content are positively correlated ($r = 0.55$), indicating that lead is partly associated with sulfides (Dai et al. 2015c). Dai (2002) also reported that the concentration of lead in pyrite within Ningxia Province coals reached 129 μg/g.

Dai et al. (2005, 2006, 2008) confirmed that lead can occur in clausthalite, which is mainly found as fracture fillings, in Chinese coals. Galena and clausthalite are rare hosts for lead in coal (Liu et al. 2013b; Swaine 2000).

4 Distribution of lead in Chinese coals

4.1 Lead distribution characteristics in different areas of China

The contents of lead in Chinese coals in different areas are presented in Table 5. Based on the collected data, the arithmetic means of lead in the coals from different provinces in China are presented in Figs. 1 and 2 and Table 5. The arithmetic mean does not accurately reflect the content of lead in coal in all provinces since it is susceptible to extreme data; therefore, the concentration coefficient CC, defined as the element concentration in the investigated coals versus a reference coal (Dai et al. 2012), was used to represent the concentration of trace elements in coals. The categories of CC were defined as: depletion ($CC < 0.5$), normal ($0.5 < CC < 2$), slightly enriched ($2 < CC < 5$), enriched (5 < CC < 10), significant enriched (10 < CC < 100), and abnormally enriched (CC > 100).

Based on the data presented in Tables 5 and 6, coals from both the Tibet–Western Yunnan and southern areas had elevated lead concentrations. Among the Chinese areas of coal distribution, the highest average lead concentration in coal was found in the Tibet–Western Yunnan area (67.2 µg/g) followed by the southern area (29.9 µg/g). These two regions contain coalfields with unusually high lead concentrations, including the Kaili coalfield (429 µg/g) (Wu et al. 2008) and the Panxian coalfield (347 µg/g) (Ren et al. 2006) in Guizhou Province, the Yong'an coalfield in Fujian Province (422 µg/g) (Lu et al. 1995), and the Qiangtang coalfields in the Tibet Autonomous Region (310 µg/g) (Fu et al. 2013). However, the highest lead content of 790 µg/g was found in a coal from the Shenfu–Dongsheng coalfield in Shaanxi Province (Ren et al. 2006). Coals from the northwestern area had the lowest lead contents, with an average value of 6.8 µg/g.

4.2 Lead contents in coals from different coal-forming periods

There were too few samples from the Late Jurassic to Early Cretaceous (J_3–K_1) period to draw conclusions as to why the arithmetic mean lead concentration was so high for these coals; there were also too few samples from the Middle Triassic (T_2) and Paleogene to Neogene (E–N) periods to comment on their lead values (Table 1). Thus, we mainly focused on the C_2–P_1, P_2, and J_{1-2}, which individually account for 38.1%, 7.5%, and 39.6% of total Chinese reserves (Dai et al. 2012), respectively, and together account for 98.4% of the samples considered in this study. As shown in Table 1, the calculated lead contents in coals from different periods decreased in the

Table 5 Coal reserves and lead contents in coals for different provinces in China

Administrative division	Coal reserves[a] (10^9 t)	Sample number	Min (µg/g)	Max (µg/g)	Arithmetic mean (µg/g)	CC
Anhui	273.6	51	5.2	347	17.3	1.0
Beijing	29.1	1	17.2	17.2	17.2	1.1
Chongqing[b]	20.5	41	1.6	92.1	17.5	1.2
Fujian	10.6	5	5.0	422	32.3	2.1
Gansu	93.1	2	2.8	13.5	8.1	0.5
Guangdong	5.8	2	21.5	24.4	23.0	1.5
Guangxi	21.8	26	4.7	59.0	21.4	1.4
Guizhou	508.0	120	3.8	429	57.1	3.8
Hebei	185.7	134	0.30	86.6	20.2	1.3
Heilongjiang	200.8	2	7.1	22.5	14.8	1.0
Henan	238.0	32	10.4	517	194	12.8
Hubei	5.0	2	22.7	53.8	38.3	2.5
Hunan	33.1	15	5.2	43.9	16.7	1.1
Inner Mongolia	2226.1	7437	< 0.20	159	13.3	0.9
Jiangsu	37.1	4	2.3	50.6	17.8	1.2
Jiangxi	14.1	14	6.6	31.0	16.4	1.1
Jilin	23.1	57	1.6	69.0	30.7	2.0
Liaoning	70.6	5	4.3	42.2	15.2	1.0
Ningxia	309.3	29	3.8	62.2	17.9	1.2
Qinghai	42.3	44	0.90	43.4	12.1	0.8
Shaanxi	1554.6	784	0.30	790	10.6	0.7
Shandong	266.8	59	5.2	36.5	17.0	0.9
Shanxi	2500.9	168	0.20	64.7	18.5	1.2
Sichuan	138.2	69	1.7	49.7	11.6	0.8
Xinjiang	1136.3	224	< 0.20	84.3	5.6	0.4
Tibet	0.9	50	11.9	310	96.3	6.4
Yunnan	240.9	68	2.8	89.6	25.2	1.7
Zhejiang	0.1	2	11.6	22.9	17.1	1.1

[a]According to Mao and Xu (1999); [b]according to Tang et al. (2006)

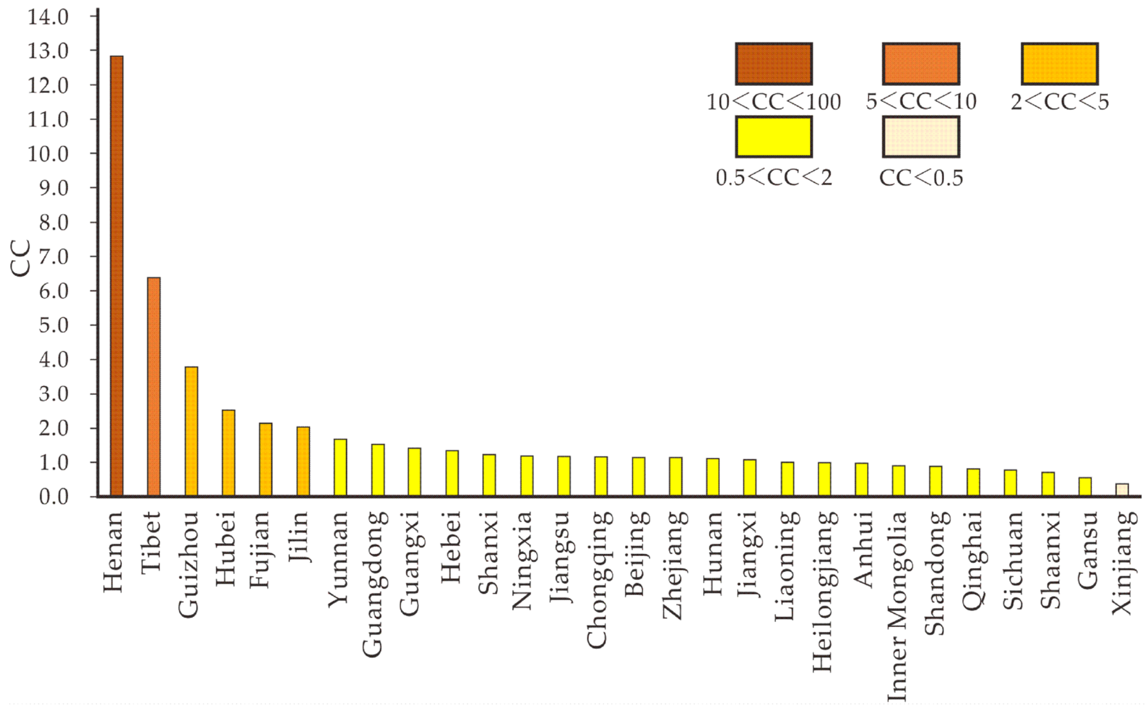

Fig. 2 CC values of lead in coals from different provinces in China. CC: Concentration coefficient. The lead value from Dai et al. (2012) of 15.1 μg/g was used as the reference value to determine the CCs. No data were available for Taiwan, Hong Kong, Macau, Hainan, Tianjin, and Shanghai

Table 6 Arithmetic means of lead contents in coals from different coal-bearing regions

Coal distribution area	Sample number	Min (μg/g)	Max (μg/g)	Arithmetic mean (μg/g)
Northeastern	7402	< 0.20	159	13.3
Northern	1326	0.20	790	19.0
Southern	357	1.6	429	29.9
Northwestern	271	< 0.20	84.3	6.8
Tibet–Western Yunnan	91	3.5	310	67.2

following order: Late Jurassic and Early Cretaceous > Late Carboniferous and Early Permian > Paleogene and Neogene > Late Permian > Late Triassic > Early and Middle Jurassic. This trend is different than that reported by Ren et al. (2006) and Tang and Huang (2004). However, the arithmetic mean content of lead in the 9447 Chinese coal samples considered in this study was 15.0 μg/g, similar to that reported by Ren et al. (2006) (15.5 μg/g). Due to the large number of samples included in this compilation along with the comprehensive analysis and corroboration of the data sources, the statistical conclusion below might be credible and would compensate for a few questionable analyses.

4.2.1 Late carboniferous to early Permian (C_2–P_1)

The content of lead in C_2–P_1 coals ranged from 0.20 to 517 μg/g with a mean of 30.3 μg/g. The lowest mean lead concentration was found in coals from the Pingshuo coalfield in Shanxi Province (0.20 μg/g) (Ren et al. 2006), while the highest was found in coals from the Pingdingshan coalfield in Henan Province (517 μg/g) (Feng et al. 2015). Based on the calculated CC values, lead was significantly enriched in coals from Henan and Guizhou Provinces and enriched in coals from Hunan and Jilin Provinces (Fig. 3).

4.2.2 Late Permian (P_2)

The content of lead in P_2 coals ranged from 1.6 to 422 μg/g with an average of 26.0 μg/g. The lowest mean lead concentration was found in coals from the Donglin coalfield in

Chongqing Municipality (1.6 μg/g) (Chen et al. 2015), while the highest was observed in coals from the Yong'an coalfield in Fujian Province (422 μg/g) (Lu et al. 1995). Overall, lead was slightly enriched in coals from Guizhou, Hebei, Ningxia, Hubei, Fujian, and Guangxi Provinces (Fig. 4).

4.2.3 *Early-middle Jurassic (J_{1-2})*

The content of lead in J_{1-2} coals ranged from less than 0.20 to 790 μg/g with an average of 12.7 μg/g. The lowest mean concentration was found in coals from the Zhundong coal mine in the Xinjiang Uygur Autonomous Region (< 0.20 μg/g) (Zhuang et al. 2013). Lead was significantly enriched in coals from the Shenfu–Dongsheng coalfield (790 μg/g) (Ren et al. 2006) and the Tongchuan coalfield in Shaanxi Province (468 μg/g) (Yang et al. 2008). Overall, lead was slightly enriched in coals from Chongqing and depleted in coals from the Xinjiang Uygur Autonomous Region (Fig. 5). At this point, we cannot explain why the arithmetic mean of lead in J_{1-2} coals was approximately half those of the Late Carboniferous to Early Permian (C_2–P_1) and Late Permian (P_2) coal samples.

5 Genetic factors affecting lead enrichment in Chinese coals

Geological factors influence the enrichment of trace elements in coals (Dai et al. 2010a). Dai et al. (2012) described five genetic enrichment types of trace elements: source rock-controlled, marine environment-controlled, hydrothermal fluid-controlled, groundwater-controlled, and volcanic ash-controlled types. The types of peat-forming plants may also affect the concentrations of trace elements in coals (Yang et al. 2008).

5.1 Source rock-controlled lead enrichment

Two processes can influence the element contents in coals from source rocks: (1) element enrichment in the land-source area and (2) the weathering or leaching of elements from the basement rocks or surrounding rocks of coal. Generally, the composition of the sediment-source region located on the margin of the coal basin is the dominant factor affecting the concentrations of trace elements in the coal basin.

We first focus on the elemental enrichment of source rocks near coal. The compositions and enrichments of elements in different types of source rocks can differ widely. Yang et al. (2008) reported that the lead concentration regularly increases through the transition from ultrabasic rock to intermediate rock and to acid rock. A comparison of the lead isotopic compositions of coals from the Upper Permian coal mines in Guizhou Province with those of different potential sources indicated that lead in these coals may come from basalt, dolomite, volcano ash, or low-temperature hydrothermal fluids (Cheng et al. 2013; Dai et al. 2012, 2017b; Mao 1991). However, tuffaceous

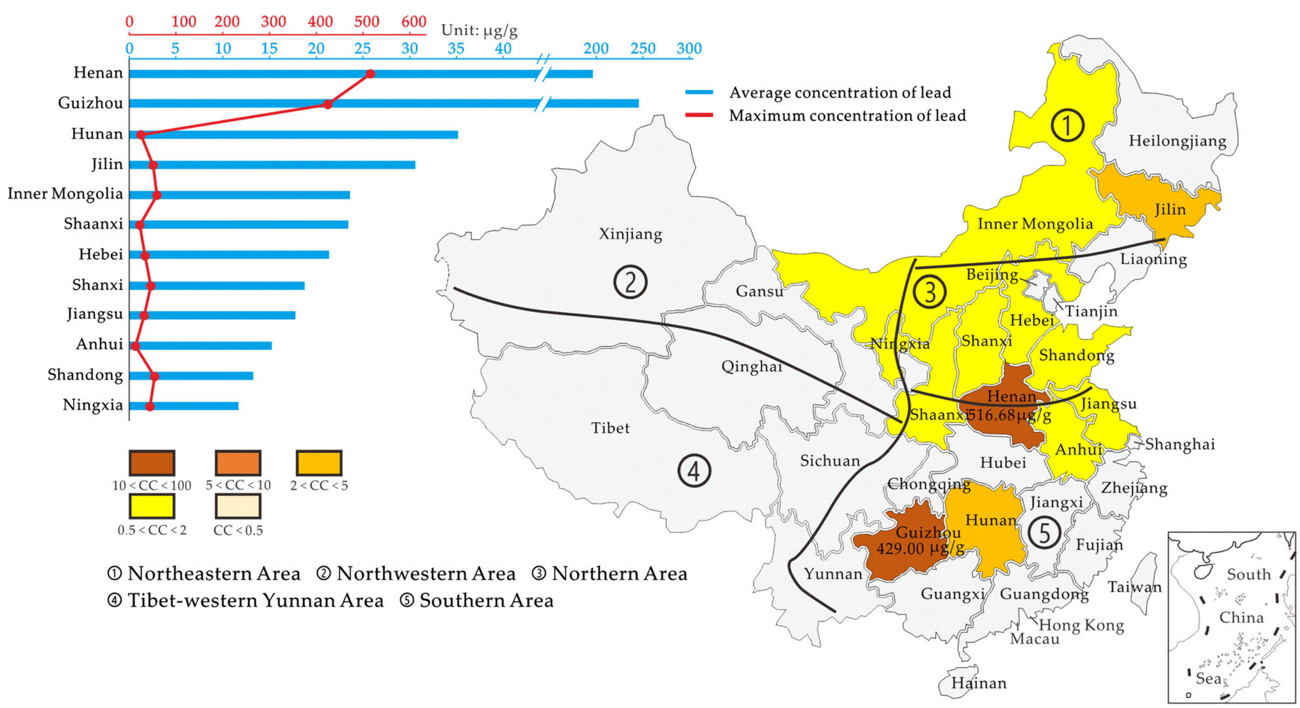

Fig. 3 Distribution of lead in Chinese coals from the late Carboniferous to Early Permian (C_2–P_1) period. CC: concentration coefficient

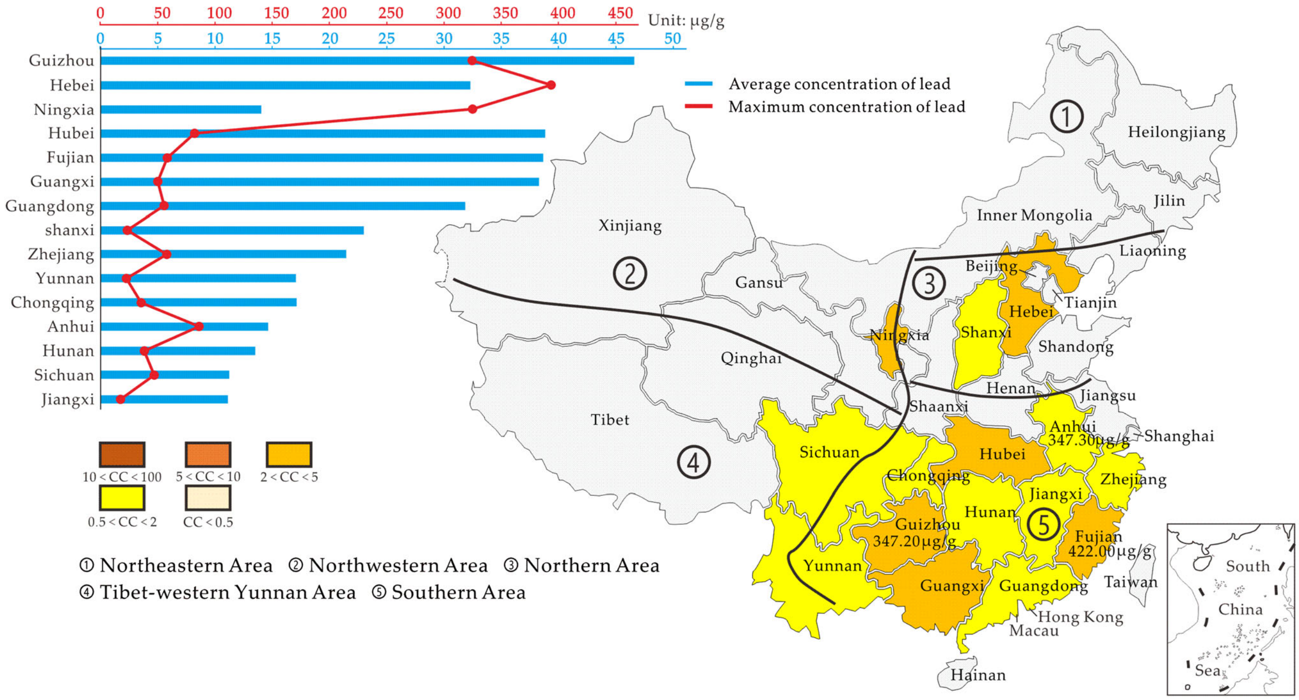

Fig. 4 Distribution of lead in Chinese coals from the Late Permian (P_2) period. CC: Concentration coefficient

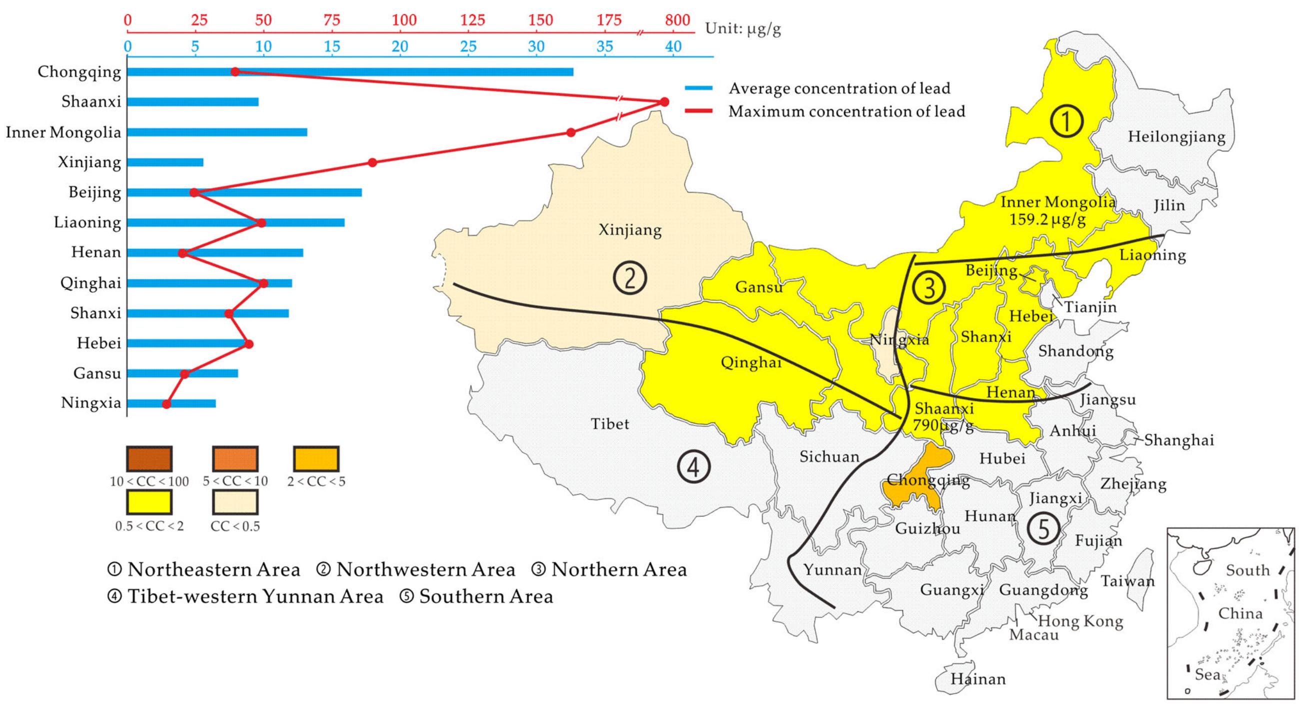

Fig. 5 Distribution of lead in Chinese coals from the Early to Middle Jurassic (J_{1-2}) period. CC: Concentration coefficient

beds in the coalfield were found to contribute little to the enrichment of lead (Sia and Abdullah 2012).

Lead can be easily weathered or leached from high-lead ores. Mihaljevič et al. (2009) reported that the lead content in coal results from a combination of lithogenic and ore lead based on a comparison of the $^{206}Pb/^{207}Pb$ ratios. Smoliński et al. (2014) found that lead in coals from the eastern zone of the Upper Silesian Coal Basin was sourced from zinc and lead ores deposited in Triassic formations. The enrichment of lead may have partly originated from the erosion or leaching of the underlying shale bed (Fu et al. 2013).

5.2 Marine environment-controlled lead enrichment

In general, the trace element content in different environments increases in the following order: freshwater < brackish water < saltwater. Seawater and streamwater usually contain very low concentrations of trace elements; the average lead contents in seawater and surface water are 0.03 and 2 μg/g, respectively (Mason and Moore 1982). It is not yet clear why seawater affects the content of elements in coal. Seawater in the peat-forming environment can change the chemical characteristics of the paleo-mire. This effect is mainly reflected in the role of bacteria in the alkaline reduction medium, which is one of the main factors affecting the enrichment of trace elements in coals (Yang et al. 2008). Wang et al. (2005) indicated that seawater intrusion in later peatization caused the kaolinite transformed to illite and montmorillonite in coals near the roof, which increased the contents of total sulfur, pyritic sulfur, sulfate sulfur and lead in coals near the roof, from Antaipu Mine, Shanxi Province.

5.3 Hydrothermal fluid-controlled lead enrichment

Hydrothermal fluid-controlled lead enrichment includes magmatic-controlled, low-temperature hydrothermal fluid-controlled, and submarine exhalation-controlled lead enrichment (Dai et al. 2012). Few previous studies have evaluated the effect of groundwater on lead enrichment in coals.

Regarding magmatic-controlled lead enrichment, Fu et al. (2013) confirmed that lead enrichment in Qiangtang Basin coals is partly related to magmatic and hydrothermal fluids. Sulfide minerals such as galena and sphalerite were observed in the Jianou coal, leading to the enrichment of lead and zinc (Dai et al. 2012).

As for low-temperature hydrothermal fluid-controlled enrichment, abnormally high contents of harmful elements in coals have been related to the hydrothermal fluids and volatiles in fault zones (Dai and Ren 2007; Dai et al. 2005, 2006; Ren et al. 2006). Epigenetic hydrothermal fluids were the major factor in the local enrichment of trace elements in coal (Dai et al. 2012). Epigenetic lead-rich minerals derived from hydrothermal fluids in coals include pyrite, ankerite, and clays. Hydrothermal pyrite, which can form during the early stages of coal formation and interact with basinal brines and hydrothermal fluids during burial, may be the main carrier of lead (Diehl et al. 2012; Li et al. 2006; Xiao et al. 2018). Lead ion is reported to be transported into coal seams through hydrothermal fluids (Guo et al. 1994). In addition, pyrite is reported to form in coal beds under the effect of tectonic deformation, allowing hydrothermal epigenetic solutions to penetrate the coal (Karayiğit et al. 2017a, b). High lead content was found in vein ankerite as a result of the influx of calcic and siliceous low-temperature hydrothermal fluids (Dai et al. 2005). Late-stage hydrothermal fluids also affect the concentration of lead in coal (Li et al. 2006; Qin et al. 2016). Dai et al. (2013b) found that episodes of epigenetic hydrothermal activity occurred after coalification and did not have a noticeable impact on lead distribution in the Fusui Coalfield in Guangxi Province; however, it resulted in the enrichment of lead in the floor rocks.

5.4 Peat-forming plant-controlled lead enrichment

The types of peat-forming plants, contents of trace elements in plants, and growth environments of plants may influence the enrichment of trace elements in coal (Yang et al. 2008). Bowen (1979) found that Cd, Co, Cr, Cu, Pd, U, and V were highly enriched in bacteria and marine algae and less so in horsetails and ferns. Wu et al. (2008) found that the enrichments of Co, Cu, and Pb were closely related in inertinite. Sun et al. (2017) indicated that lead generally has an affinity for vitrinite, and its origin is associated with the parenchymatous and woody tissues of roots, stems, barks, and leaves, which are composed of cellulose and lignin. However, this enrichment mechanism is generally less important than the other mechanisms discussed above.

6 Conclusions

The arithmetic mean of lead concentration in Chinese coals was determined in this study to be 15.0 μg/g. Taking the Chinese coal reserves into consideration, the weighted-average lead concentration was calculated as 19.6 μg/g, slightly higher than the arithmetic mean for world coal. The maximum lead concentration in Chinese coals was 790 μg/g in the Shenfu–Dongsheng coalfield in Shaanxi Province.

Lead was significantly enriched in the coals from Henan Province and enriched in the coals from the Tibet Autonomous Region. Coals from the Tibet–Western Yunnan and southern areas showed elevated lead concentrations compared to coals in other regions. Lead was enriched in the coals from northern China formed during the Late Carboniferous to Early Permian period. Lead was also enriched in coals formed during the Late Permian period in northwestern China, the southeastern Tibet–Yunnan area, and the southern area.

Sulfides, generally pyrite and galena, are the primary hosts of lead in Chinese coals; however, lead in coals is also associated with silicates, organic matter, carbonates, clays, and other minerals. Organic-bound lead was identified in low-rank coals.

Source rocks supply most of the elements during the coal-forming process and influence the mineral components and element concentrations in coals. Marine water had a considerable impact on lead accumulation in coal by affecting the chemical characteristics of peat swamp systems. Hydrothermal fluids and peat-forming plants also influenced the enrichment of lead in some coalfields in China.

While lead poisoning has caused a range of serious human health problems, no known environmental or human health problems in China have been attributed to the mobilization of lead from coal combustion. Moreover, it appears that most coals in China have modest levels of lead. Nevertheless, it would be wise to minimize the dispersal of lead into China's environment. This can be accomplished by minimizing the use of high-lead coals, discouraging the domestic use of coal without proper ventilation, expanding the use of coal-cleaning techniques to remove mineral-associated lead prior to combustion, and using efficient post-combustion pollution control systems.

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Compliance with ethical standards

Conflict of interest There is no conflict of interest regarding the publication of this research article.

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