Chromium in Chinese coals: geochemistry and environmental impacts associated with coal-fired power plants

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Abstract Chromium (Cr), one of the prime hazardous trace elements in coals, may engender adverse effects on eco-environment and threaten human health during utilization of coal. Based on the samples obtained in our laboratory and published literature, the abundance and modes of occurrence of Cr in Chinese coals, and the environmental impacts associated with coal-fired power plants (CFPPs) were elucidated in this study. With a total of 1397 sets of data, the mean concentration of Cr in Chinese coals was calculated as 21.33 μg/g by the “reserve-concentration” weighted calculation method. Spatially, the average Cr contents increased gradually from North China to South China. Temporally, coals from T3, E–N and P2 were relatively enriched in Cr compared to the other geological time. The Cr concentration in coal varied with different coal ranks. The geological factors accounted for Cr enrichment in coals could be divided into the primary, secondary and epigenetic processes. Higher percentages of organically Cr occurred in low-rank coals, while inorganically associated Cr was mainly found in clay minerals. After coal combustion, most of Cr was enriched in solid wastes (e.g., fly ash and bottom ash). The leaching of Cr from solid wastes in the rainy season (especially acid rain) needs to be a concern for CFPPs. It was estimated that the atmospheric emission of Cr from CFPPs increased annually from 2015 to 2019 and reached approximately 159 tons in 2019.

Keywords Chromium · Geochemistry · Chinese coals · Environmental impact · Coal-fired power plants

Introduction

Although the COVID-19 crisis completely reshaped the global coal market, China was still the largest coal producer and consumer in the world (IEA, 2020). Compared with the statistics in 2009, China’s coal production and consumption increased by 24.0% and 15.7% in 2019, respectively (BP, 2020). Despite the
adjustment of energy structure and industrial transformation policy to fulfill the goals of "carbon–neutral" by 2060 domestically, coal is still and will continue to be significant energy for power generation in the future (Mallapaty, 2020). Coal combustion in coal-fired power plants (CFPPs) was a main anthropogenic source of hazardous trace elements (HTEs) entering the environment (Ostad-Ali-Askari, 2022; Turan, 2021, 2022; Zheng et al., 2017).

Chromium (Cr) was one of the key HTEs in coals (Dai et al., 2012a). It was a variable-valent metal element and usually occurred as Cr(III)/Cr(VI) in the environment (Javadinejad et al., 2019; Shah et al., 2012). Compared with Cr(III), Cr(VI) had strong carcinogenicity to human body (Velma et al., 2009). The International Agency for Research on Cancer (IARC) has designated Cr(VI) compounds as human carcinogens (IARC, 2020). Long-term intake of Cr had adverse effects on human reproduction, and might cause lung cancer, adenocarcinoma, flat epithelial cancer, etc. (Stanley et al., 2014). Cr released into ecosystem could accumulate in organisms, e.g., bacteria, plants and aquatic life, eventually enriching through the biological chain to threaten human health (Pushkar et al., 2021).

China's coal resources were rich in reserves and diverse in coal types (Jie et al., 2020). The geological settings of different coalfields varied greatly, resulting in wide Cr concentration ranges and complex modes of Cr occurrence in Chinese coals (Ren et al., 2006). The combustion of high Cr coals can bring potential occupational health problems, public disease risks and environmental deterioration (Finkelman et al., 2019). Qin et al. (2018) demonstrated that the mode of occurrence of Cr had an important impact on the Cr volatilization during coal combustion, thus affecting the distribution of Cr in combustion by-products. The secondary release of Cr could happen during the disposal of combustion solid wastes (e.g., bottom ash and fly ash) (Fu et al., 2019). Moreover, the long-term interaction of Cr with surface water/groundwater systems in outdoor emplacements has attracted considerable attention (Ostad-Ali-Askari et al., 2016; Zhao et al., 2020). Liu et al. (2012) reported that there was an incident of Cr leaching from fly ash contaminated surface water in Guizhou Province, China, which attracted widespread attention. Accordingly, the stack emission of fine particulate-bound Cr and leaching of solid wastes were probably the most significant concerns for Cr contamination from CFPPs.

To date, the systematic geochemical information of Cr in Chinese coals was relatively limited. Cr was recommended for priority monitoring in CFPPs emissions by the U.S.A Electric Power Research Institute (Goodarzi, 2006). However, there was no specific emission standard for Cr emitted from CFPPs in China. In this study, our laboratory conducted experiments on Cr in Chinese coal samples and combined with abundant reported literature to comprehensively elucidate the geochemistry of Cr and the environmental impacts of Cr associated with CFPPs in China. The main aims included the following: (1) the spatial–temporal distribution of Cr in Chinese coals, (2) the possible geological reasons for the abnormal enrichment of Cr in coals, (3) the modes of occurrence of Cr, and (4) the partitioning, leachability and atmospheric emission inventory of Cr in Chinese CFPPs. This research can provide meaningful references for the database of Cr in Chinese coals and will contribute to environmental management as well as the emission standard revision.

Materials and methods

Data collection

A total of 1397 sets of data related to Cr in coals were compiled from 28 provinces, cities and autonomous regions in China. Among them, 304 coal samples in our laboratory were collected from 11 provinces of China (including Anhui, Shandong, Hubei, Sichuan, Chongqing, Liaoning, Xinjiang, Shanxi, Hebei, Inner Mongolia and Yunnan Provinces). These coal samples were used to analyze the concentration and speciation of Cr, and the corresponding experimental methods were shown in “Elemental determination” section and “Sequential chemical extraction procedure” section. Additionally, the data obtained in the published literature on Cr concentrations in coals from different coal-producing regions of China were also used to supplement. Through careful inspection of the data sources, the duplicate data were effectively eliminated. The data could accurately represent Cr concentrations, geographic information and geological background in Chinese coals.
Elemental determination

Approximately 0.1 g of each pulverized coal sample (<200 mesh) was digested by HNO₃ (5 mL), HF (5 mL) and HClO₄ (3 mL) in the Teflon digestion vessels. The samples were then heated for 9 h in a muffle furnace at 180 °C. After cooling down, the samples were heated on an electric hot plate at 160 °C for 4 h for acid-driving. Subsequently, the digested products were diluted to 100 mL with ultrapure water, and then shaken well. The concentrations of Cr in the diluent were determined by inductively coupled plasma mass spectrometry (ICP-MS).

Sequential chemical extraction procedure

To investigate the modes of occurrence of Cr, Cr in coal samples was extracted and categorized as six phases by the sequential chemical extraction (SCE) procedure. The main steps of the six-step SCE procedure are shown in Fig. 1. According to the procedure illustrated in Fig. 1, Cr in coals could be operationally partitioned into water leachable, ion exchangeable, organic matter bound, carbonate bound, silicate bound and sulfide bound in proper sequence. The Cr in each fraction was analyzed by ICP-MS. Coal standard reference material SARM20 was used to evaluate the accuracy of Cr determination. The analytical value for Cr was within the certified values, with recovery rates ranging from 85.3 to 106.2%.

Spatial-temporal distribution of Cr in Chinese coals

Cr content in Chinese coals

Previous literature data on Cr content in Chinese coals

As one of the important HTEs in coals, the abundance and distribution of Cr in Chinese coals have been studied since the late 1960s (Wu et al., 2005). The average concentration of Cr in Chinese coals is summarized in Table 1. Zhao (1997) analyzed 137 Chinese coal samples and obtained an...
average Chinese Cr value (34.9 μg/g), with the range of 0.46–942.7 μg/g. Tang et al. (2002) gave an average Cr abundance of 12.0 μg/g on the basis of 1410 Chinese coal samples. Wu et al. (2005) estimated an average Cr content in Chinese coals was 17.2 μg/g based on 1367 samples. Ren et al. (2006) took the reserve weight in different coal-forming periods into consideration, and obtained an average Chinese Cr content (15.3 μg/g) based on 1601 samples. Bai et al. (2007) reported that the concentration of Cr in Chinese coals was 16.94 μg/g based on 1123 coal samples, which was slightly higher than the value (15.4 μg/g, n = 1615) from Dai et al. (2012a). The arithmetic mean of Cr concentration in Chinese coals was calculated to be 31.98 μg/g based on 1048 available field-test data by Tian et al. (2013). Cao et al. (2021) obtained the mean concentration of Cr was 14.7 μg/g from 875 samples according to their database management system of Chinese coals. These research data could complement the basic geochemical data of Cr in Chinese coals. Overall, it can be seen that there was slight difference among different literature-reported values. A more accurate result which reveals the level of Cr content in Chinese coals can be obtained with new research data by using reasonable calculation method.

**Updates and evaluation of Cr contents in Chinese coals** This paper removed several extreme values of Cr content in individual coal mines from the data source and assigned weights on the basis of coal reserves from different coal-producing regions in China. Based on the experimental data from our research and combined with the data available in published literature, this study calculated Cr content from 1397 coal samples of the twenty-eight coal-producing provinces, cities and autonomous regions in China (Table 2). The concentration of Cr in Chinese coals ranged from 0.012 to 971.0 μg/g, with an arithmetic mean of 29.37 μg/g. The calculation method for evaluating the mean concentrations of HTEs in coals was particularly critical, because the uneven distribution of coal samples and several super-high Cr areas could cause inaccuracies of the final result (Kang et al., 2011). When the relative proportion of coal reserve from different regions was taken into account, it could be more scientifically to reflect the average abundance of Cr in Chinese coals (Ren et al., 2004). The weighting factor was the percentages of coal reserves of each coal-producing region in China, and this paper adopted the data from China Statistical Yearbook 2017. Accordingly, by using the “reserve-concentration” weighted calculation method, the mean concentration of Cr in Chinese coals was calculated as 21.33 μg/g. The mean concentration of Cr in Chinese coals was higher than that in Australia (10.0 μg/g), Indonesia (7.40 μg/g), Russia (18.0 μg/g) and USA (15.0 μg/g) and significantly lower than that from India coal (32.03 μg/g) (Finkelman, 1993; Kong et al., 2007; Liu et al., 2015a, 2015b; Shah et al., 2012; Yuan et al., 2013). Compared to the world’s

### Table 1: Cr concentrations in coals from various countries in the world

| Countries | Number of samples | Range (μg/g) | Average (μg/g) | References |
|-----------|------------------|-------------|---------------|------------|
| China     | 137              | 0.46–942.7  | 34.87         | Zhao (1997) |
| China     | 1410             | 2.0–50      | 12.0          | Tang et al. (2002) |
| China     | 1367             | 0.10–422    | 17.18         | Wu et al. (2005) |
| China     | 1601             | 0.10–942.7  | 15.33         | Ren et al. (2006) |
| China     | 1123             | Nd          | 16.94         | Bai et al. (2007) |
| China     | 1615             | Nd          | 15.4          | Dai et al. (2012a) |
| China     | 1048             | 0.02–971.0  | 31.98         | Tian et al. (2013) |
| China     | 875              | 0.02–971.0  | 14.7          | Cao et al. (2021) |
| Australia | Nd               | Nd          | 10.0          | Shah et al. (2012) |
| India     | 113              | 2.0–195     | 32.03         | Kong et al. (2007) |
| Indonesia | 31               | 1.1–24.9    | 7.40          | Liu et al. (2015a, 2015b) |
| Russia    | Nd               | Nd          | 18.0          | Yuan et al. (2013) |
| USA       | 7847             | Nd          | 15.0          | Finkelman (1993) |
| Worldwide | 236              | 0.50–60     | 16.0          | Ketris and Yudovich (2009) |

Nd: no data
Table 2  Abundance of Cr in coals from different provinces, cities and autonomous regions of China

| Province | Number of samples | Minimum (μg/g) | Maximum (μg/g) | Arithmetic mean (μg/g) | Coal reserve (100 mt) | References |
|----------|------------------|----------------|----------------|------------------------|----------------------|------------|
| Anhui    | 337              | 3.24           | 201.00         | 25.65                  | 82.37                | Huang et al. (2001), Yang et al. (2004), Chen et al. (2006), Wu et al. (2005), Kong et al. (2007), Chen et al. (2009), Chen et al. (2010), Tian et al. (2013), This research |
| Beijing  | 1                | 6.00           | 6.00           | 6.00                   | 2.66                 | Ren et al. (2006) |
| Chongqing| 41               | 5.92           | 185.73         | 28.48                  | 18.03                | Li et al. (2006), Ren et al. (2006), Zhuang et al. (2007), Dai et al. (2010), This research |
| Fujian   | 3                | 8.40           | 52.70          | 30.48                  | 3.98                 | Lu et al. (1995), Ren et al. (2006) |
| Gansu    | 3                | 16.47          | 40.87          | 23.70                  | 27.32                | Tian et al. (2013) |
| Guangdong| 2                | 2.00           | 74.00          | 38.00                  | 0.23                 | Ren et al. (2006) |
| Guangxi  | 38               | 11.20          | 971.00         | 115.26                 | 0.90                 | Lu et al. (1995), Zeng et al. (2005), Ren et al. (2006), Song et al. (2007b) |
| Guizhou  | 138              | 0.02           | 167.30         | 25.62                  | 110.93               | Zhao et al. (1997), Ni et al. (1998), Zeng et al. (1998), Zhuang et al. (2001), Dai et al. (2006a), Chen et al. (2006), Dai et al. (2006b), Li et al. (2006), Yang (2006), Song et al. (2007b), Dai et al. (2010), Tian et al. (2013) |
| Hebei    | 51               | 3.79           | 73.60          | 30.91                  | 43.27                | Ren et al. (2006), Tian et al. (2013), This research |
| Heilongjiang | 19             | 0.02           | 46.00          | 14.66                  | 62.28                | Wang et al. (1996), Querol et al. (1997) |
| Henan    | 15               | 3.42           | 68.30          | 24.78                  | 85.58                | Lu et al. (1995), Lv et al. (2003), Chen et al. (2006), Ren et al. (2006), Tian et al. (2013) |
| Hubei    | 28               | 3.00           | 201.00         | 46.12                  | 3.20                 | Lu et al. (1995), Ren et al. (2006), Zhuang et al. (2007), This research |
| Hunan    | 14               | 2.88           | 173.00         | 37.08                  | 6.62                 | Wang et al. (1999), Ren et al. (2006), Tian et al. (2013) |
| Inner Mongolia | 113         | 2.10           | 119.00         | 13.32                  | 510.27               | Wang et al. (1996), Zhuang et al. (2006), Ren et al. (2006), Dai et al. (2006), Yang (2008), Dai et al. (2012b), This research |
| Jiangsu  | 10               | 7.80           | 42.00          | 18.75                  | 10.39                | Ren et al. (2006), Zhou et al. (2011), Tian et al. (2013) |
| Jiangxi  | 19               | 9.78           | 134.00         | 39.74                  | 3.36                 | Querol et al. (2001), Ren et al. (2006) |
| Jilin    | 11               | 4.12           | 95.27          | 27.23                  | 9.71                 | Wang et al. (1996), Querol et al. (1997), Wu and Zhou (2004) |
| Liaoning | 15               | 3.10           | 145.00         | 28.63                  | 26.73                | Wang et al. (1996), Querol et al. (1997), Kong et al. (2001), Liu and Zhang (2006), Ren et al. (2006), Gao and Yang (2008), This research |
| Ningxia  | 14               | 2.20           | 52.00          | 10.77                  | 37.45                | Ren et al. (2006), Song et al. (2007b) |
| Qinghai  | 4                | 23.85          | 43.94          | 30.82                  | 12.39                | Ren et al. (2006) |
| Shaanxi  | 23               | 3.75           | 108.00         | 32.36                  | 162.93               | Ren et al. (2006), Tian et al. (2013) |
| Shandong | 103              | 0.25           | 112.00         | 19.34                  | 75.67                | Lu et al. (1995), Yang et al. (2004), Chen et al. (2006), Jiang et al. (2011), Tian et al. (2013), This research |
The concentration of Cr in coal (16.0 μg/g) reported by Ketris and Yudovich (2009), slightly high level of Cr concentration was observed in Chinese coal.

Spatial distribution of Cr in Chinese coals

**Abundance of Cr in coals from different regions in China** The relative enrichment of HTEs in coals could be evaluated by concentration coefficient (CC), which was expressed as follows (Dai et al., 2014):

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CC = \frac{\text{element concentration in investigated coal samples}}{\text{element concentration in world hard coals}}
\]

(1)

where 6 categories were designated: depleted (CC<0.5), normal (0.5<CC<2), slighted enriched (2<CC<5), enriched (5<CC<10), significantly enriched (10<CC<100) and unusually enriched (CC>100). The concentration of Cr in world hard coals was 17 μg/g (Ketris & Yudovich, 2009), which was used as the denominator for the normalization.

The spatial distribution of Cr in Chinese coals from different provinces, cities and autonomous regions is visually shown in Fig. 2. According to the CC values, coals with mean Cr concentrations that reached the “enriched” (5<CC<10) and “slight enriched” (2<CC<5) grades were approximately distributed in southern China and central China. As intuitively shown in Table 2 and Fig. 2, the Cr-rich regions in southern China were Guangxi Province (115.26 μg/g), Yunnan Province (70.95 μg/g), Tibet (47.43 μg/g) and Guangdong Province (38.00 μg/g). The central Cr-rich regions were mainly concentrated in Hubei Province (46.12 μg/g) and Hunan Province (37.08 μg/g). The mean concentrations of Cr in eastern provinces were attributed to “normal” (0.5<CC<2) for the most part, except Jiangxi Province (39.74 μg/g). The Cr concentration in coals from northern China was relatively low, and Beijing (6.0 μg/g) and Xinjiang Province (7.91 μg/g) were categorized as “depleted” (CC<0.5). Furthermore, the mean Cr concentration in coals from Ningxia Province (10.77 μg/g), Inner Mongolia (13.32 μg/g), Heilongjiang Province (14.66 μg/g) and other provinces was also lower than the mean concentration of Cr in Chinese coals calculated from this study. In conclusion, the abundance of Cr in coals increased discretely from northern China to southern China.

**Distribution of Cr in coals from high-Cr areas** Abnormally high values of Cr content in coal mines were from some regions of China. The content of Cr in high-organic-sulfur coals was up to 389 μg/g.
in Ganhe coalmine (Yunnan Province) and 408 μg/g in Chenxi Coalfield (Hunan Province), respectively (Li et al., 2013; Zhao et al., 2017a, 2017b). The Cr content in stone coal sample from Enshi (Hubei Province) was two orders of magnitude higher than the national average of Cr in coals, reaching up to 1738 μg/g (Liu et al., 2016). Dai et al. (2017) reported that Cr was extraordinarily enriched in the Late Permian coals from the Moxinpo Coalfield of Chongqing, with the range from 1984 to 2899 μg/g. In addition, Cr was also extremely enriched in coals from Baise and Heshan Coalfields in Guangxi Province, Guiding Coalfield in Guizhou Province, Huabei Coalfield in Anhui Province and Shenbei Coalfield in Liaoning Province, with the maximum concentration of 118 μg/g, 269 μg/g, 1094 μg/g, 101 μg/g and 145 μg/g, respectively (Dai et al., 2013, 2014; Ren et al., 2004; Yan et al., 2019; Zheng et al., 2008). In this study, the Cr concentrations in 189 Late Permian coal samples from the Xieqiao coal mine, Huainan City of Anhui Province, were determined, and the mean concentration of Cr was up to 416.15 μg/g, with a range of 37.24–740.23 μg/g.

Temporal distribution of Cr in Chinese coals

The abundance of Cr in Chinese coals was highly correlated with different coal-forming periods (Dai et al., 2012a). The coal-forming ages of China could be sorted into six periods, which included Carboniferous and Permian (C–P), Late Permian (P2), Late Triassic (T3), Early Jurassic and Middle Jurassic (J1–2), Late Jurassic and Early Cretaceous (J3–K1), and Eocene and Neogene (E–N) (Ren et al., 2006). As

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**Fig. 2** The spatial distribution of Cr in coals from different provinces, cities and autonomous regions of China. CC: a ratio of Cr investigated coal samples versus world hard coals.
shown in Fig. 3 and Table 3, with the data based on definite coal-forming periods from our data source, the average Cr concentration in Chinese coals of different geological periods increased in the following order: $J_{1-2}$ (12.67 $\mu$g/g) < $J_{3-K_1}$ (13.27 $\mu$g/g) < C–P (16.28 $\mu$g/g) < $P_2$ (30.74 $\mu$g/g) < E–N (45.56 $\mu$g/g) < $T_3$ (49.31 $\mu$g/g). Through the Kruskal–Wallis test, the Cr concentration in coals among the coal-forming periods had extremely significant difference ($\chi^2 = 58.74, \rho = 0.00$). The total predicted coal reserves in China were made up of 64.7% $J$ coal, 21.5% C–P coal, 8.7% $P$ coal, 0.5% $T_3$ coal and the proportion of coals in other ages was less than 5% (Chen et al., 2021a, 2021b). Therefore, although Cr abundance in Chinese coals from $T_3$, E–N and $P_2$ was relatively high, the reserves of such high-Cr coals were low. Additionally, $T_3$ and E–N coals were mainly distributed in Tibet-western Yunnan area, and $P_2$ coals primarily located in Southern area (Dai et al., 2012a). To improve the certainty and representativeness of the evaluation of Cr abundances in Chinese coals, more coal samples from $T_3$, $J_3-K_1$ and E–N are needed.

**Cr abundance in coals of different ranks**

Coal rank is a very important parameter that can affect the coal utilization in industrial processes, and up to now, low-rank coal pyrolysis technology still suffers from low economic efficiency, difficult tar quality control and wastewater treatment (Chang et al., 2016; Ostad-Ali-Askari & Shayannejad, 2021). Bouška and Pešek (1999) reported an arithmetic mean of Cr in 4219 lignite samples around the world was 54.99 $\mu$g/g. Valkovic (1983) calculated the mean value of Cr in global bituminous coals to be 10.0 $\mu$g/g (Ren et al., 2006). The mean Cr content in anthracite of the world coals was estimated to be 17.0 $\mu$g/g by Юдoвич and Кeтpиc (2005). Based on our data source, the abundance of Cr from different coal ranks in China is shown in Table 4. The arithmetic mean of lignite, bituminous coal and anthracite was 18.02 $\mu$g/g ($n=107$), 19.87 $\mu$g/g ($n=912$) and 23.65 $\mu$g/g ($n=248$), respectively. It was obvious

| Geological period                  | Number | Range (μg/g) | Arithmetic mean (μg/g) |
|------------------------------------|--------|--------------|------------------------|
| Carboniferous and Permian (C–P)    | 521    | 0.25–68.30   | 16.28                  |
| Late Permian ($P_2$)               | 209    | 2.00–543.54  | 30.74                  |
| Late Triassic ($T_3$)              | 52     | 8.00–134     | 49.31                  |
| Early Jurassic and middle Jurassic ($J_{1-2}$) | 109 | 0.46–78.3    | 12.67                  |
| Late Jurassic and early cretaceous ($J_3-K_1$) | 40  | 3.00–45.5    | 13.27                  |
| Eogene and Neogene (E–N)           | 56     | 2.90–189     | 45.56                  |
that the concentration of Cr gradually increased from lignite through bituminous coal to anthracite. Some high-rank coals in southern China were influenced by complex geological factors that might lead to the enrichment of Cr in coals, as we would discuss below.

Possible reasons for the distribution of highly enriched Cr in Chinese coals from geological perspectives

As shown in “Spatial distribution of Cr in Chinese coals” section, many coalfields with extremely high concentrations of Cr are located nationwide, particularly in southern China. The enrichment of Cr in coal was the comprehensive result of various geological factors, and these factors could be divided into three stages, which contained primary, secondary and epigenetic processes (Dai et al., 2012a). The primary factors played a major role in the formation period of peat mire, mainly including the properties of parent rock in the sediment-source region, sedimentary environment, and coal-forming plants (Liu et al., 2001). The secondary factors impacted the migration and enrichment of Cr in the stage of peat coalification (lithification), which were mainly related to the fluid injection process and synsedimentary volcanic activity, etc. (Liu et al., 2021). The epigenetic factors mainly referred to the material exchange of Cr in coals after the formation of coal seam, for instance, tectonic movement, groundwater and oxidation/weathering processes (Golian et al., 2020; Ren et al., 2006). The enrichment of Cr in coals might be the result of superposition of the above-mentioned diversified factors.

The primary factors The properties of parent rock in the sediment-source region located on the margin of coal basin had a strong influence on the background values of Cr in coals (Ren et al., 2006). The sediment-source region was a dominant factor in Cr enrichment in some fault-controlled basins or coalfields, because the Cr-rich detrital materials could be transported into the interior of coal basin (Dai et al., 2012a). For instance, the Southwest Coal Basin (SCB) was a fault-controlled coal basin which covered a relatively vast area of Southwest China, including western Guizhou, eastern Yunnan, southern Sichuan and Chongqing (Dai et al., 2017; Li et al., 2016; Wang et al., 2012; Zhang et al., 2004b; Zhao et al., 2015). The sediment-source region of SCB was the Kangdian Oldland, which was mostly made up of basaltic rocks and led to the enrichment of the Cr in considerable part of Late Permian coals (Dai et al., 2012a). Besides, the Beipiao Coalfield was a small intracontinental basin in Liaoning Province, and average Cr content in the Jurassic coal from this coalfield was as high as 55.82 μg/g, which was mainly due to the Cr-rich basaltic eruption in the basement (Ren et al., 2006). Driven by strong oxidation and weathering processes in the supergene zone, Cr in parent rocks could be dissolved, transported and finally entered the coal accumulation basin (Dai et al., 2011).

Coal-forming plants was another factor determining the background value of Cr in coals. The concentrations of Cr in lower plants, such as marine algae and mosses, were generally higher than that in higher plants. For instance, the Cr concentration in the ash of bryophytes was eight times higher than that of higher plants (Vassilev et al., 1997). The Cr concentration in sapropelic coals was obviously higher than that in humic coals, which was because sapropelic coals were mainly formed by algae or herbaceous plants while humic coals were mainly formed by higher plants (Liu et al., 2003). The coal forming plants of P₂ and T₃ coals were relatively low-grade plants (mainly Gigantopterids and fern flora), which might account for the relatively high Cr content in coals of these two coal-forming periods mentioned above. On the contrary, the flourished coal-forming plants in Jurassic period were advanced plants (gymnosperm), which might contribute to the relatively low Cr content in Jurassic coals (Cao et al., 2021).

In addition, the redox potential, pH and seawater invasion in sedimentary environment were also important factors contributing to the Cr enrichment in coals. The content of Cr was low or absent in the sediments of oxidizing environment, but Cr tended to be enriched in the sediments of reducing environment (Tribovillard et al., 2006). Cr could form soluble chromate and migrate under strong oxidizing conditions, whereas insoluble Cr compounds would be formed and then easy to be enriched in humic acid environment which was dominated by reduction (Hu et al., 2014). Acidic medium was favorable for the migration of Cr(III), but Cr tended to form Cr(OH)₃ precipitates in alkaline environments (Tan, 2019). The Cr concentration in seawater (0.2–60 μg/L) was much higher than that in freshwater (0.1–6.0 μg/L) (Ren et al., 2006). Compared with Shanxi Formation
of Late Paleozoic from North China, the Cr concentration in Taiyuan Formation of the same provenance area was relatively high, which was chiefly affected by seawater and coastal swamp (Liu et al., 2001).

Secondary and epigenetic factors The intrusion of magmatic hydrothermal fluids from synsedimentary volcanic activities within the coal basin could provide rich sources for Cr in coals. (Dai et al., 2003a, 2003b, 2005a; Finkelman et al., 1998; Querol et al., 1997). Our previous study has demonstrated that the Cr content reached the enrichment level in Wolonghu coalmine (Anhui Province), which was due to the effect of frequent magmatic hydrothermal during Mesozoic and Cenozoic in eastern China (Chen et al., 2021a, 2021b). Hydrothermal fluids played an important role in the local enrichment of Cr from Late Permian coals in southern China, e.g., Zhijin and Qinglong Coalfields in Guizhou Province (Dai et al., 2005b). Moreover, M9 coals of Yanshan Coalfield from Southeast Yunnan Province were extremely rich in Cr (329 μg/g), which was the result of joint action by metal-rich hydrothermal solutions from submarine injection and synsedimentary volcanic ash (Dai et al., 2010). Magmatic hydrothermal fluids accelerated the coalification process, and with the deepening of coal metamorphism, the relative content of organic-Cr in coals decreased sharply (Zhou et al., 2011).

The epigenetic factors affecting the enrichment of Cr in coals mainly included late tectonic movement, groundwater migration and weathering (Liu et al., 2001; Ostad-Ali-Askari et al., 2019). Magmatic activity after coal formation could cause changes in the structure of the material within the coal seam, and re-enrichment of Cr in coals occurred (Ren et al., 2006). The heat generated during tectonic movement could also affect the metamorphic degree of coal seam (Zhang et al., 2000). Except for the magmatic hydrothermal solution, epigenetic fluids which included groundwater could enter the coal seams through fissures or fault structures (Dai et al., 2012b). The metal ions and microbial effects which were rich in groundwater might increase the concentration of Cr in coals (Eslamian et al., 2018; Liu et al., 2001). In addition, Goodarize et al. (1987) suggested that the content of Cr in weathered coals was relatively high. Currently, due to the unique high oxidation characteristics, weathered coal was mainly used as a source of humic acid and fertilizer to improve soil properties (Semenova & Patrakov, 2017). Nonetheless, the study about the influence of weathering on the Cr content in coals was scarce, so new and detailed investigations were required.

Modes of occurrence of Cr in coals

The speciation of HTEs in coals can be classified into inorganic/mineral, organic and intimate organic associations (Dai et al., 2020, 2021). Due to the limitations of direct methods (microanalytical techniques), indirect methods which include statistical analyses (mainly consisting of correlation analysis, cluster analysis, and principal components analysis), density separation experiments, and SCE have been the prime options for exploring the affinities and modes of occurrence of HTEs in coals (Dai et al., 2021; Kang et al., 2011; Qin et al., 2018). The current consensus is that Cr in coals can be attached not only in inorganic mineral form, but also in organic form (Dai et al., 2021; Finkelman et al., 2002; Qin et al., 2015; Swaine, 1998; Tian et al., 2013). The relevant researches were summarized below and are demonstrated in Fig. 4.

Inorganically associated Cr

Association with clay minerals

To date, it was well known that the inorganic Cr in coals mainly occurred in clay minerals (Huggins et al., 2000a; Qin et al., 2015). Due to the large specific surface area and various surface functional groups, clay minerals had a strong adsorption capacity for Cr (Huang et al., 2021). As early as 1981, Finkelman analyzed American coals by using scanning electron microscope and energy dispersive spectrometry (SEM–EDS), and found Cr was related to clay minerals (Finkelman, 1981). Huggins et al. (2000b) tested the mode of occurrence of Cr in American bituminous coals by using X-ray absorption fine structure (XAFS) spectroscopy, SEM and electron microprobe analysis (EMPA), and found Cr was related to clay minerals (Finkelman, 1981). Huggins et al. (2000b) tested the mode of occurrence of Cr in American bituminous coals by using X-ray absorption fine structure (XAFS) spectroscopy, SEM and electron microprobe analysis (EMPA), and demonstrated that 10–50% of the total Cr associated with illite. For the No.8 coal seam in Qinglong mine from western Guizhou Province, Cr was abnormally enriched (154.2 μg/g) and mainly associated with clay minerals according to SCE experiments (Li & Tang, 2005). Wang et al. (2011) studied Ga-rich coal in Jungar Coalfield (Inner
Mongolia) by SEM–EDS and X-ray diffraction spectrometer (XRD), and found that Cr mainly occurred in the clay minerals (principally illite). Results of the cluster and factor analysis on the coal samples from Huainan and Huaibei Coalfields (Anhui Province) showed that Cr occurred in clay minerals (Chen et al., 2009; Shao et al., 2015). Finkelman et al. (2018) made concluding comments, indicating that HF leachable Cr was certainly associated with clay minerals, and CH₃COONH₄ leachable Cr was possible in exchangeable sites in clays. Additionally, Cr could also exist in clay minerals in the form of isomorphism (Yang et al., 2020). By using SCE methods, Zhao (1997) determined Cr occurrences in bituminous coal and anthracite in China, and observed that most Cr occurred in clay minerals (94.9–100%), which might be due to the isomorphic substitution of Cr(III) and Al(III). Moreover, Wang et al. (2019a, 2019b) concluded that the presence of humic acid could increase the adsorption of Cr on clay minerals.

**Association with other minerals**

Cr-bearing minerals in coals mainly include spinel group minerals (such as chromite, magnetite and chromian magnetite), pyroxenes, crocoite, garnets and amphiboles, etc. (Finkelman et al., 2019). Cr in coals can also occur in sulfides and can replace Fe(III) and Al(III) in the form of Cr(III) into silicate minerals during the magmatism process (Ren et al., 2006). In China, strong correlation between Cr and Al in Huayingshan coals (Sichuan Province) demonstrated the affinity of Cr with aluminosilicate (Zhuang et al., 2012). Gao et al. (2015) elaborated that Cr in coals from Huolinhe mine (Inner Mongolia) was significantly and positively correlated with pyrite, quartz and carbonate minerals through float/sink methods and correlation analysis. For the high-organic-sulfur coal of Heshan Coalfield, Guangxi Province, Shao et al. (2015) found that Cr had a strong correlation with Fe and a weak correlation with Al; thus, they suggested that Cr in the coals might be bound with sulfides or phosphates. Besides, compared with Heshan Coalfield, Cr in high-organic-sulfur coal of Chenxi Coalfield (Hunan Province) showed a positive correlation with Fe as well as a significant positive correlation with Al, suggesting that Cr in these coals mainly occurred in pyrites (Li et al., 2013). According to our recent electron probe microanalysis (EPM) analysis of bituminous coal from Zhaotong (Yunnan Province), it was found that in addition to clay minerals, Fe-minerals were the main carrier of Cr in this coal (Tang et al., 2022a).

Organically associated Cr

Experiments have demonstrated that Cr in coals had an organic affinity (Swain et al., 1992; Huggins et al., 2000a; Huggins & Huffman, 2004; Dai et al., 2005b, 2013; Finkelman et al., 2018, 2019). Huggins and Huffman (2004) found that Cr from some American bituminous coals occurred as nano-sized particles of Cr-oxy-hydroxides (CrOOH) in the organic matter through XAFS and demonstrated that organically associated Cr in coals could be largely shielded from attack by acidic reagents. Zhao et al. (2003) and Ren et al. (2004) reported that Cr had a strong organic affinity in low-rank coals from Ordos coal basin and
Shenbei Coalfield. Their results also showed that the average organic fraction of Cr in coals was about 20%, which was mainly associated with humic acid, fulvic acid and/or organic macromolecular compounds. In Yanshan and Guiding Coalfields, Yunnan and Guizhou Provinces, Cr was mainly associated with organic matter in superhigh-organic-sulfur coals (Liu et al., 2015a, 2015b). Through further nanoscale mineralogical examination, Zhao et al. (2017a, 2017b) clarified that Cr in super-high-organic-sulfur coal from Ganhe coalmine (Yunnan Province) was occurred in organic components by using transmission electron microscopy (TEM) technique. Nonetheless, organic occurrence of Cr in coals was not readily leached by reagents (Riley et al., 2012). Finkelman et al. (2018) indicated that there was no CH\textsubscript{3}COONH\textsubscript{4} leachable Cr in bituminous coals, and the leaching behavior of Cr in the low-rank coals was about 40% silicate, 5% sulfide, 10% oxide, and 5% organic association. The relative concentration of organic Cr in coals decreased sharply with the increase of coal metamorphism, and most of high-rank coals did not contain organically associated Cr (Zhang et al., 2000). The influence of coal rank on organic occurrence Cr was complex, and the mechanism for this has not been elucidated.

On the basis of comprehensive investigation of the modes of occurrence of Cr in coals, the occurrence forms of Cr in lignite from Lincang (Yunnan Province), bituminous coal from Daizhuang (Shandong Province) and anthracite from Zhaotong (Yunnan Province) have been tested by SCE procedure in our laboratory. The fractions of different forms of Cr in coal samples are shown in Fig. 5. Cr in coals by this six-step solvent extraction method could be operationally defined as water leachable, ion exchangeable, carbonate-bound, silicate-bound, sulfide-bound and organic matter-bound. The concentration of Cr in the lignite, bituminous coal and anthracite samples was 9.28, 12.48 and 40.70 μg/g, respectively. The concentrations of water-leachable Cr in these feed coals were insignificant and even below detection limit in anthracite. From low-rank coal to high-rank coal, the proportions of Cr combined with carbonate bound matter in coals increased remarkably, while the proportions of Cr existed in ion-exchangeable and silicate-bound forms decreased gradually. The sulfide-bound Cr in anthracite was lower than in bituminous coal and lignite. The organic matter-bound Cr was converted to other forms in the course of metamorphism of coals. Most of all, the Cr partitioning in different occurrence forms might be closely related to the metamorphic degree of coals.

**Fig. 5** Fraction of Cr in selected coal samples defined by sequential extraction experiment
Environmental impacts of Cr associated with CFPPs

CFPPs were one of the most important anthropogenic emission sources of Cr into the environment (Tian et al., 2014). The environmental problems associated with CFPPs which included the accumulation of solid wastes, pollution of ambient soil/groundwater and emission of air pollutants after power generation had attracted sustained attention (Tauqeer et al., 2022a, 2022b; Turan et al., 2017). Clear understanding of the partitioning and emission characteristics of Cr in CFPPs was helpful to study the environmental impact of Cr.

Partitioning of Cr in solid wastes from CFPPs

Minerals and organic fractions would undergo thermal decomposition, fusion, disintegration and agglomeration during coal combustion, and then, Cr in coals can be liberated and distributed into solid wastes (Tang et al., 2018). The volatility of Cr affected its degree of volatilization from coal and the partitioning in solid wastes (Tang et al., 2022b). Following the explication by Clarke (1993) and Meij et al. (2004), Cr in coals could be categorized as an element between nonvolatile and semi-volatile and was mainly retained in solid wastes after coal combustion. Researchers have reported that air pollution control devices (APCDs) in CFPPs had remarkable impacts on the partitioning of Cr in solid wastes (Li et al., 2020). This paper compared the Cr concentrations in different solid wastes from CFPPs in China over recent years, and presented them in detail in Table 5. As shown in Table 5, compared with the feed coal, Cr was enriched in the fly ash and bottom ash after coal combustion, whereas existed less in the flue gas desulfurization (FGD) gypsum. For most CFPPs, a higher Cr concentration in fly ash could be observed, and the reasons for this phenomenon were described as follows: (1) in comparison with bottom ash, fly ash had smaller particle size and stronger adsorption capacity for the condensation of Cr (Han et al., 2021; Tang et al., 2013, 2022b), (2) the occurrence of Cr in coals was an important influencing factor for its redistribution in solid wastes, and Cr in the form of organic/sulfide combinations was more likely to partially evaporate and thus coagulate on the fly ash (Fu et al., 2019; Li et al., 2020). In summary, the solid wastes from CFPPs needed to be carefully disposed to prevent their possible mobilization into the environment.

Table 5  Cr concentrations in the feed coal (FC), bottom ash (BA), fly ash (FA) and FGD gypsum (FG) from different CFPPs in China

| Boiler type | APCDs | Installed capacity (MW) | FC (μg/g) | BA (μg/g) | FA (μg/g) | FG (μg/g) | Reference |
|-------------|-------|-------------------------|-----------|-----------|-----------|-----------|-----------|
| PC          | ESP + WFGD | Nd                      | 26.53     | 66.28     | 85.53     | Bdl       | Tang et al. (2013) |
| PC          | SCR+ESP+WFGD+WESP | 660       | 12.30     | 47.90     | 76.15     | 8.94      | Zhao et al. (2016) |
| PC          | SCR+ESP+WFGD+WESP | 660       | 12.8      | 48.0      | 87.0      | 28.0      | Zhao et al. (2017a, 2017b) |
| PC          | SCR+ESP+WFGD | 300       | 1.16      | 66.40     | 37.54     | 5.83      | Zhang et al. (2017) |
| PC          | SCR+ESP+WFGD | 600       | 22.55     | 29.75     | 95.14     | 14.10     | Fu et al. (2019) |
| PC          | SCR+FF+FGD | 350       | 12.17     | 33.31     | 43.96     | Nd        | Chen et al. (2019) |
| CFB         | SCR+FF+FGD | 110       | 24.89     | 54.72     | 41.85     | Nd        | Chen et al. (2019) |
| CFB         | SCR+FF+FGD | 75        | 7.58      | 30.67     | 27.32     | Nd        | Chen et al. (2019) |
| PC          | SCR+ESP+WFGD+WESP | 300       | 14.50     | 52.10     | 76.90     | 8.94      | Wang et al. (2019a, 2019b) |
| CFB         | SCR+ESP-FF+WFGD | 2 × 300   | 97.0      | 187.1     | 195.1     | 38.9      | Li et al. (2020) |
| PC          | SCR+ESP+WFGD | 4 × 600   | 69.0      | 192.4     | 155.1     | 36.6      | Li et al. (2020) |
| CFB         | SCR+ESP-FF+WFGD | 3 × 200   | 96.0      | 198.6     | 213.5     | 31.2      | Li et al. (2020) |
| PC          | SCR+LTT-ESP+WFGD | 350       | 20.80     | 37.30     | 53.20     | 12.40     | Han et al. (2021) |

Bdl: below determination limit; Nd: no data; PC: pulverized coal boiler; CFB: circulating fluidized bed boiler; SCR: selective catalytic reduction; ESP: electrostatic precipitator; LTT: low-low temperature; FF: fabric filter; FGD: flue gas desulfurization; WFGD: wet flue gas desulfurization; WESP: wet electrostatic precipitator
The leachability of Cr in solid wastes

The storage activities of solid wastes produced by CFPPs might cause environmental problems because the leachable Cr polluted groundwater/surface water and soil (Wang et al., 2020). The leaching amount of Cr was closely related to the content of Cr in solid wastes and external leaching conditions (e.g., liquid-to-solid ratio, leaching time and pH value) (Zhang et al., 2019). Zhao et al. (2018a) determined the leaching behavior of Cr in fly ash from CFPPs and found that the Cr content in fly ash was directly proportional to the quantity of Cr leached. Reardon et al. (1995) reported that the leaching of Cr showed an obvious relationship with the ratio of water to the coal ashes from CFPPs. As the cumulative liquid-to-solid ratio increased and the leaching time continued, the leaching concentration of Cr in bottom ash and fly ash increased at the beginning and reached the maximum at a certain liquid-to-solid ratio, and then decreased (Zhang et al., 2019). Under natural conditions, the contact of the bottom ash, fly ash and gypsum with water produced alkaline leachates, but the leaching capacity of Cr under alkaline conditions was much lower than under acidic conditions (Zhao et al., 2020). Besides, the leachability of Cr was also related to the speciation (Fu et al., 2019). Cr can easily dissolve in its hexavalent state into leachate, including chromates and dichromates, whereas trivalent Cr was less soluble (Huggins & Huffman, 2004). Almost all Cr in coal existed in relatively benign trivalent form, but the combustion process would convert some of Cr(III) into a more toxic and carcinogenic hexavalent (Cr(VI)) (Shah et al., 2012). During coal combustion, the organically bound Cr was easily oxidized into Cr(VI) than the mineral-bound Cr (Tang et al., 2022a). Fortunately, the proportion of Cr(III) in the solid wastes was absolutely superior compared to Cr(VI) (S’wietlik et al., 2014). Moreover, Fu et al. (2019) carried out leaching experiments and concluded that Cr in exchangeable and carbonate bound fractions were easier to leach than in residual form. Thus, CFPPs should scientifically manage the solid wastes and pay attention to the runoff flow during the rainy season (especially acid rain), so to prevent Cr contaminated water or sediments from entering the environment or even the food chain.

Atmospheric Cr emissions of CFPPs in China

Although the removal efficiency of APCDs in CFPPs had reached a relatively high level, part of Cr in the stack gas could still escape to the atmosphere and cause potential hazards (Han et al., 2021; Li et al., 2020). Identifying the atmospheric Cr emissions from CFPPs was inherent in improving air quality and human health. In this article, a method for calculating the atmospheric emissions (AE) of Cr from CFPPs in China was adopted (Chen et al., 2013):

\[
AE = C \times c_i \times AER
\]

where \(C\) was the annual coal consumption by CFPPs in China, which was derived from the data of China Statistical Yearbook 2021. And \(c_i\) was the mean concentration of Cr in Chinese coals, which was taken from the value of 21.33 μg/g in this study. \(AER\) was the atmospheric emission ratio of Cr in CFPPs, which was the geometric mean of the reported values in China (Fig. 6). From the data of previous research (Fig. 6), the atmospheric emission ratio of Cr in Chinese CFPPs showed a variation from 0.03 to 6.41%. In this study, the atmospheric emission ratio of Cr incorporated in the calculation was 0.355%, which was between the atmospheric emission ratio of CFPPs in Japan (0.42%) and the reported value of a Canadian CFPP (0.17%) (Huggins & Goodarzi, 2009; Ito et al., 2006).

The atmospheric Cr emissions of CFPPs in China from 2015 to 2019 are presented in Table 6. It could be perceived that the atmospheric emission of Cr caused by Chinese coal-fired power generation increased annually from 2015 to 2019 and reached 159.0 tons in 2019. During these five years, the Cr stacks emission fluxes increased by approximately 23.1 tons, with moderate fluctuations in growth rates. At present, the research on the atmospheric emission of Cr from CFPPs was still relatively limited, more detailed filed data needed to be determined, to improve the data statistics of Cr pollution and provide support for issuing corresponding emission limits of Cr.

Conclusions

Based on 1397 sets of Chinese coal samples data, the concentration of Cr in Chinese coals was between
The reserve-weighted mean concentration of Cr in Chinese coals was calculated to be 21.33 μg/g, which could reduce error due to the uneven distribution of samples from different areas. There were significant variations in the distribution of Cr in coals from different coal-producing regions in China. Overall, the abundance of Cr in coals in South China was distinctively higher than that in North China. The Cr concentration in Chinese coals increased in the following order: J1–2 < J3–K1 < C–P < P2 < E–N < T3. With the increase of coal rank, the concentration of Cr also increased gradually. The enrichment of Cr in coals was the result of diversified geological factors, and these factors could be divided into three stages: primary, secondary and epigenetic processes. In addition, Cr in coals can occur in both inorganic and organic forms, and the most common inorganic form of Cr was associated with clay minerals. As a nonvolatile or less volatile element, Cr tended to be enriched in solid wastes (fly ash and bottom ash) after coal combustion from CFPPs. Due to the leaching characteristics of Cr, it was recommended that CFPPs need to pay special attention to the management of solid wastes during the rainy season, especially during the acid rain period. The inventory of atmospheric emissions of Cr from CFPPs in China from 2015 to 2019 was also estimated. This paper can provide scientific references for public health evaluation and environmental policy formulation.

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