

Chemical Characteristics and Utilization of Coal Mine Drainage in China

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Abstract The growing demand for water in China's coal-producing regions requires enhanced coal mine drainage (CMD) utilization. Mine water quality was analyzed for 269 mines distributed in 11 of China's large coal-producing regions. We found that China's CMD can be broadly characterized as: slightly contaminated, acidic, high salinity, high sulfate, high fluoride, and containing elevated iron and manganese. When CMD had properties of more than one category, its most distinctive characteristic was used for classification. When this was done, the chemical characteristics tended to correlate with the hydrogeological conditions of the region. Appropriate treatment technologies and pollution prevention measures based on these chemical characteristics could enhance the likelihood of mine water being used to relieve China's water shortage crisis and promote environmental protection for China's coal-producing regions.

Keywords Coal mine drainage · Water quality · Water treatment

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Introduction

China has the world's third largest coal reserves and is first with respect to coal production and consumption. Coal production and consumption account for 76 and 69 %, respectively, of China's primary energy (Ju et al. 2009). Total coal production and consumption in 2001 was 1.16 and 1.05 billion tonnes (t), increasing to 3.66 and 3.82 billion t in 2012, respectively, which is three times what it was 11 years ago. Even though China has started developing alternative energy technologies, coal will be its major energy source for a long time.

Coal resources are unevenly distributed in China, with much of it in the north and west, with a few deposits in the south and east. Most of the coal resources occur in less developed areas, such as Shanxi, Inner Mongolia, Shaanxi, Xinjiang, Guizhou, and Ningxia. These provinces have 833.4 billion t of coal reserves, accounting for 81.8 % of China's total reserve. East and southeast Chinese provinces such as Beijing, Tianjin, Shanghai, Shandong, Jiangsu, Zhejiang, Fujian, and Guangdong require large amounts of coal while their reserves stand at 553 million t, only 5.4 % of China's total coal reserve.

The water resources of China are likewise not well-distributed; the north holds 80 % of the total coal reserves but only 20 % of its total water (Sun et al. 2012; Yuan and Shi 2008). Most of the large coal-producing regions in China, except for Yunnan-Guizhou, Huainan-HuaiBei, and eastern Inner Mongolia, suffer from severe water shortages. This is especially true for Shanxi, Ningdong, and Shandong. Additionally, with nearly a quarter of the world's population, but just 8 % of its freshwater. China's water resources only total 2,300–2,700 m³ per capita, which is one-fourth of the world average (Liu and Liao 2007). However, because 97 % of China's coal is produced by

Table 1 CMD quality statistical data (in mg/L, except for pH) of 11 large coal-producing regions in China; number of samples in brackets; minimum and maximum values in parentheses, with the median value provided afterwards, where it was felt that it would be useful

| Region and no. of investigated mines | pH | F | Fe | Mn | TDS | SO ₄ | Cl | References |
|---|----------------|-------------------------------|---------------------------|------------------------------|-------------------------------|-----------------------------|-----------------------------|--|
| Huaibei–Huainan <i>N</i> = 34 | [34] (6.8–8.9) | [14] (0.05–3.15) 0.57 | [3] (0.01–0.29) 0.23 | | [30] (430–6,084) 1,826 | [30] (1–4,285) 253 | [30] (31–1,465) 156 | Li et al. (1999), Yan et al. (2004) |
| SW Shandong <i>N</i> = 65 | [61] (2.7–9.0) | [15] (0.19–2.92) 0.91 | [17] (0–691.02) 0.50 | [2] (0.02–21.42) 10.72 | [52] (403–5,266) 1,617 | [40] (74–3,410) 1,180 | [34] (22–3,207) 104 | Chang et al. (2001), Chen and Zhang (2010), Fan et al. (2011), Li (2008), Wan et al. (2004), Wang (2000), Yin et al. (1997), Zhang et al. (2000) |
| Shanxi <i>N</i> = 23 | [22] (3.4–8.8) | [16] (0.20–5.93) 0.59 | [14] (0.03–102.9) 0.22 | [7] (0.02–8.10) 0.65 | [19] (464–4,144) 642 | [19] (3–2,726) 135 | [13] (21–225) 83 | Ge (2007), Sun et al. (2007), Wu (2008), Xu et al. (2007), Zhao et al. (2007) |
| Eastern Inner Mongolia <i>N</i> = 34 | [34] (6.3–8.4) | [29] (0–3.69) 0.86 | [28] (0.01–11.28) 0.3 | [31] (0–0.71) 0.12 | [25] (356–4,233) 1,100 | [15] (61–406) 216 | [15] (26–352) 111 | Dong et al. (2007), Huang et al. (2010), Li et al. (2012) |
| Yunnan–Guizhou <i>N</i> = 40 | [39] (2.3–8.8) | [17] (0.11–1.50) 0.39 | [37] (0.09–667) 3.00 | [23] (0.01–32.0) 0.50 | [14] (504–2,932) 1,334 | [16] (46–2,280) 725 | [14] (2–284) 5 | Cao (2007), Dong et al. (2012), Wang et al. (2008), Xu (2011) |
| Shandong <i>N</i> = 10 | [7] (7.6–8.6) | [1] (4.13) 4.13 | [2] (0.20–6.50) 3.35 | [1] (0.16) | [4] (980–1,684) 1,383 | [5] (149–651) 251 | [6] (40–287) 222 | Kou et al. (2011), Liu et al. (2013), Xiu and Zhu (2009) |
| Jizhong <i>N</i> = 9 | [8] (6.8–8.4) | [7] (0.03–0.6) 0.2 | [7] (0–1.23) 0.04 | [7] (0–0.39) 0.03 | [9] (200–5,356) 602 | [8] (7–1,818) 59 | [8] (3–1,575) 8 | Dai (2012), Zhang (2004) |
| Henan <i>N</i> = 27 | [19] (7.2–8.8) | [23] (0.3–4.59) 0.9 | [11] (0.09–32.10) 0.87 | [9] (0.01–2.35) 0.04 | [24] (350–1,965) 662 | [25] (5–643) 2,159 | [19] (5–74) 30 | Huang and Chen (2011), Li et al. (2006), Xiao and Wei (2008), Xu (2009), Zhai et al. (2010), Zhang and Yin (2010) |
| Ningdong <i>N</i> = 9 | [9] (5.6–8.2) | [1] (0.8) | [7] (0.01–100) 1.56 | [1] (10.00) | [8] (46–9,982) 4,896 | [2] (884–2,123) 1,504 | [2] (420–2,509) 1,465 | Liu et al. (2005), Xu (2010) |
| Xinjiang <i>N</i> = 7 | [7] (7.3–8.2) | [3] (0.26–0.8) 0.32 | [4] (0–0.59) 0.17 | [3] (0–0.06) 0.05 | [6] (758–22,225) 16,098 | [7] (8–4,654) 2,604 | [6] (72–9,500) 6,475 | Ablizi (2004), Gu et al. (2009) |
| Huanglong <i>N</i> = 6 | [6] (7.0–8.4) | [1] (1.6) | [1] (0.10) | [5] (240–1,450) 537 | [5] (45–243) 224 | [5] (3) | [1] (27) | Guo (2011), Xi (2011) |
| Others <i>N</i> = 5 | [5] (2.0–3.6) | [3] (159.8–287.8) 270.0 | [3] (6.1–16.5) 11.3 | [2] (2,212) | [1] (1,610–4,306) 1,951 | [3] (0–1) 0.5 | [2] (0–1) 0.5 | Bao (2001), Qin et al. (2001) |

underground mining, the pumping and discharge of large quantities of mine water is generally required for miner safety. However, the resultant contamination is now recognized as both a serious pollution problem and a waste of water resources.

Coal mine drainage (CMD) is typically contaminated by coal and rock dust, and can be used after treatment for industrial production, ecological, and domestic purposes to effectively alleviate water shortage problems. However, in China, CMD chemical characteristics differ greatly between the different regions (Feng et al. 2004; Gu et al. 2009; Li et al. 2006; Liu et al. 2013; Sun et al. 2007; Zhao et al. 2007). Previous work has mostly focused on single mining regions. As a result, it is difficult to gain the national perspective on CMD quality that is needed to assist decision making by the government. This perspective is necessary because different types of CMD require different water treatment technologies. We collected and analyzed a total of 73 CMD samples and obtained data for an additional 196 samples from 11 large coal-producing regions from mining companies and from the published literature. Based on this data (summarized in a supplementary table that will accompany the on-line version of this paper), we classified the CMD to provide a basis for district planning and management, anticipating that water research professionals and others simply interested in learning more about the CMD within their area will benefit.

Chemical Characteristics of CMD in China

Coal mine drainage (CMD) can contain a variety of contaminants (Banks et al. 1997; Cravotta 2008a, b; Plumlee et al. 1999; Rose and Cravotta 1998). Others have found that chemical characteristics of CMD tend to differ depending on an area's hydrogeological conditions (Brady et al. 1998; Cravotta and Ward 2008; Geidel and Caruccio 2000; Lottermoser 2010). In China, CMD is normally classified as: slightly contaminated, high salinity, acidic, or based on their contaminant levels (Hu 1998; Shan 1999).

Table 1 shows the CMD quality statistical data of 269 mines in 11 large coal-producing regions and other coal mining areas. The data indicates that China's CMD can be divided into six types: slightly contaminated, high salinity, acidic, high sulfate, high fluoride, and containing elevated levels of iron and manganese. The levels that we used to define these categories are provided in the last row of Table 2. Since CMD can contaminate water that can be used for drinking, these classification standards of CMD make reference to the Standards for Groundwater Quality, Standards for Surface Water Quality, and Standards for Drinking Water Quality (Table 2).

Table 2 Water quality standards related to major pollution substances in CMD and the classification standards for CMD in China (in mg/L, except for pH)

| Standards | pH | TDS | F | SO ₄ | Fe | Mn |
|--------------------------|---------|--------|------|-----------------|------|------|
| Drinking water | 6.5–8.5 | ≤1,000 | ≤1.0 | ≤250 | ≤0.3 | ≤0.1 |
| Groundwater (III level) | 6.5–8.5 | ≤1,000 | ≤1.0 | ≤250 | ≤0.3 | ≤0.1 |
| Surface water | 6–9 | – | ≤1.0 | ≤250 | ≤0.3 | ≤0.1 |
| Classification standards | >6 | ≤1,000 | ≤1.0 | ≤250 | ≤0.3 | ≤0.1 |

Table 3 Approaches for utilization of CMD in China

| Approach | Details | % of all utilized CMD ^a |
|--|--|------------------------------------|
| Industrial use coal-to-chemicals, etc. | Coal production, coal washing, coal coking, etc. | 70 |
| Ecology, agriculture | Mine area greening, dust resistance, farm irrigation, etc. | 15 |
| Domestic supply | Drinking, bathing, cleaning, etc. | 10 |

^a According to the mine water utilization development planning of China

There are some correlations between these types of CMD. For instance, acidic CMD often contains higher levels of sulfate, iron, and manganese. In addition, sulfate is often the major component in highly saline CMD. But because of its wide distribution, heavy impact on environment, and different treatment methods, high sulfate CMD and CMD with elevated concentrations of iron and manganese were separated based on their dominant characteristic. In this study, the fluoride concentrations of 46 CMD samples exceeded groundwater quality standards, posing great potential harm to human health. So, this type of CMD was singled out. Some CMD in a few mining areas contains elevated levels of trace contaminants, such as Hg, Cr, As, and Pb, but because the number of such sites in China is low, this type of CMD was not a focus of this study. The sampling distribution is shown in Fig. 1.

Slightly Contaminated CMD

This type of CMD is widely distributed in all of China's large coal-producing regions. This water usually contains pollutants such as suspended solids (SS), chemical oxygen demand (COD), oil, and small amounts of organic contaminants. The characteristics of this type of CMD are high SS, low concentrations of trace contaminants, circumneutral pH, and grey or black color. SS comes mainly from the

Fig. 1 The distribution of water samples for 269 coal mines in China

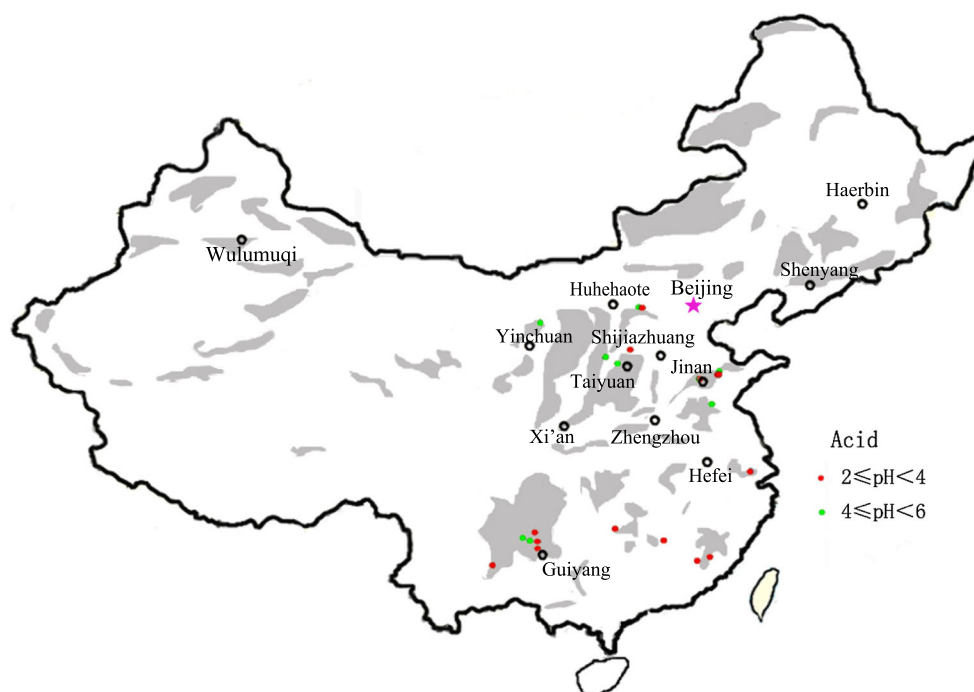
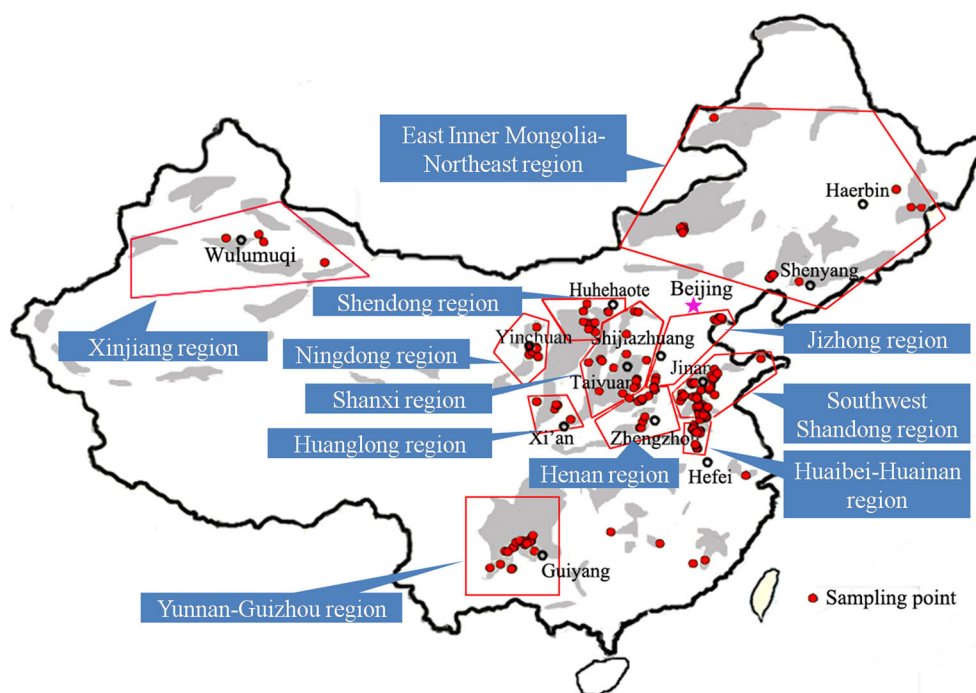


Fig. 2 Distribution of acidic CMD in China

coal and rock dust produced during mining. COD, oil and small amounts of organic contaminants come from mining equipment, human excrement, etc. This kind of CMD, with its relatively light environmental influence and simple and inexpensive treatment requirements, was widespread.

Acidic CMD

This type of CMD has pH values below 6 and generally between 2 and 4. As shown in Fig. 2, acidic CMD is mainly distributed in three large coal-producing regions located in

southwest Shandong, Yunnan-Guizhou, and Shanxi. In addition, there are a few other mining areas with scattered acidic CMD, such as Longyong of Fujian province, Changguang of Zhejiang province, Tianhe of Jiangxi province, and Jinzhushan of Hunan province. Notably, the CMD in Yong'an of Fujian province has a pH as low as 2.0.

In China, the formation of acidic CMD is generally due to the varying amounts of sulfur, with mass fractions between 0.3 and 5.0 % found in the coal seam, mainly pyrite (FeS_2) (Wang 2010; Yin et al. 2008). This type of CMD often contains elevated concentration of iron and manganese, as well as other trace metals.

Fig. 3 Distribution of CMD with high salinity in China

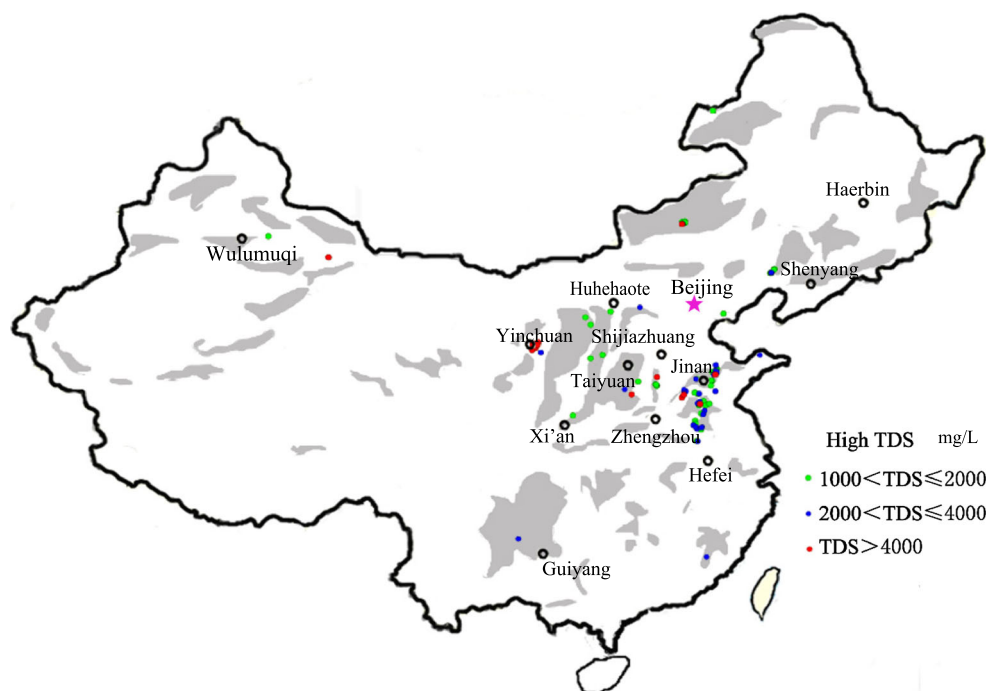
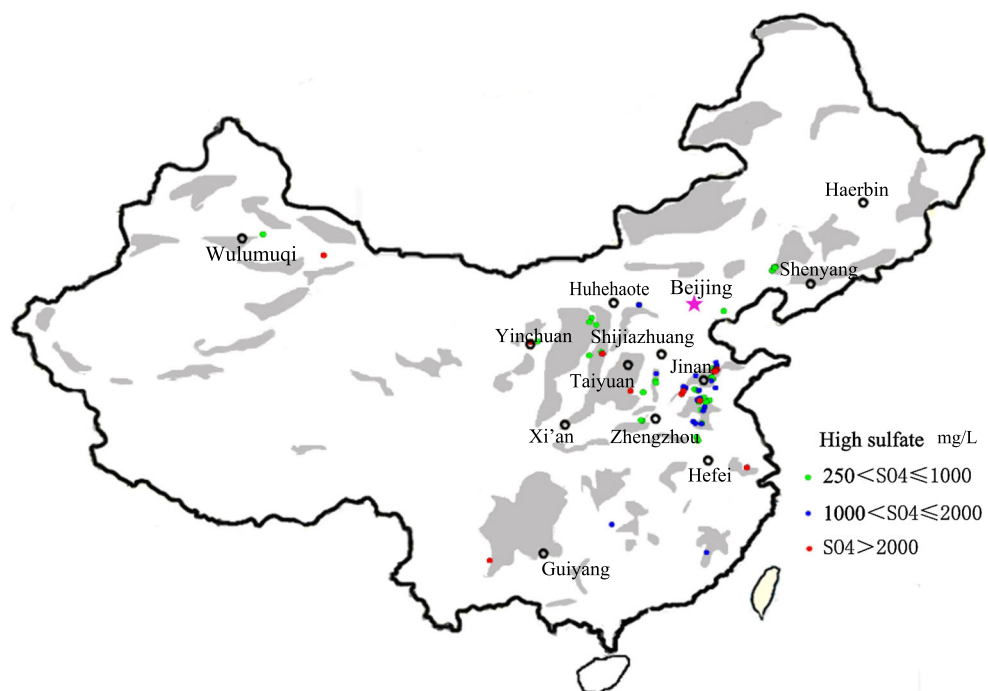


Fig. 4 Distribution of CMD with high sulfate in China



CMD with High Salinity

This CMD has salinity (SO_4^{2-} , Cl^- , Ca^{2+} , Mg^{2+} , K^+ , Na^+ , HCO_3^- , etc.) concentrations greater than 1,000 mg/L and usually has neutral or alkaline pH and a bitter taste. The samples had salinities between 45 and 22,225 mg/L, with a median of 1,197 mg/L. The more saline water was mainly distributed in Huainan-Huaipei, southwest Shandong, Shanxi,

Eastern Inner Mongolia, Jizhong, Ningdong, and Xinjiang (Fig. 3). In coalfields such as Zibo and Juye of southwest Shandong and Jincheng of the Shanxi region, the salinity concentration can be greater than 2,000 mg/L, and in the Dananhu mine of the Xinjiang region, the salinity reaches 22,225 mg/L.

Highly saline CMD is mainly formed by sulfide oxidation and the resultant free acid reacting with carbonate and

Fig. 5 Distribution of CMD with high fluorine in China

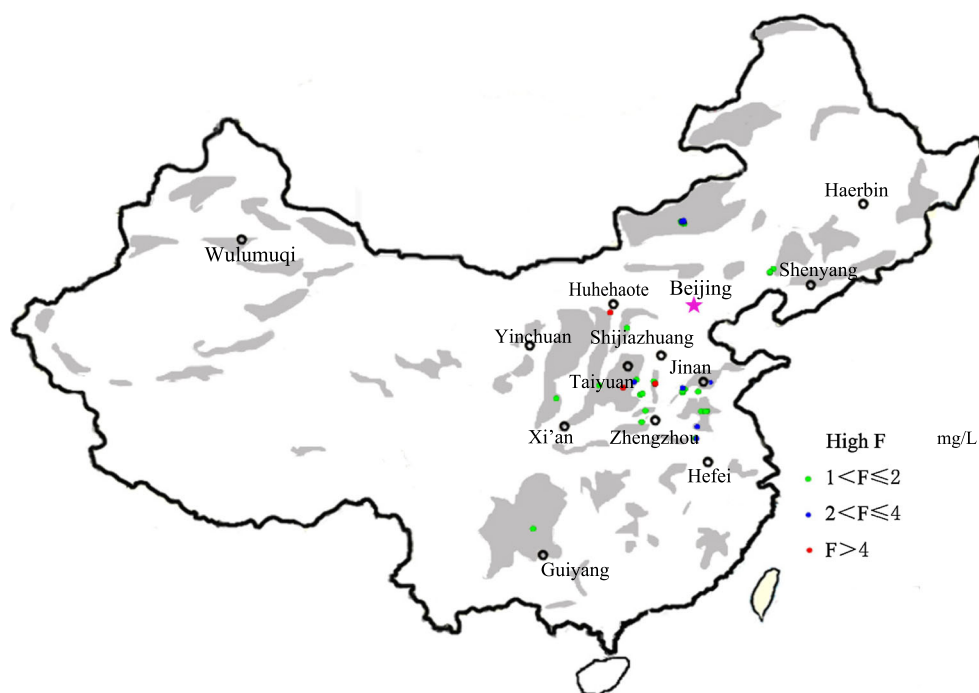
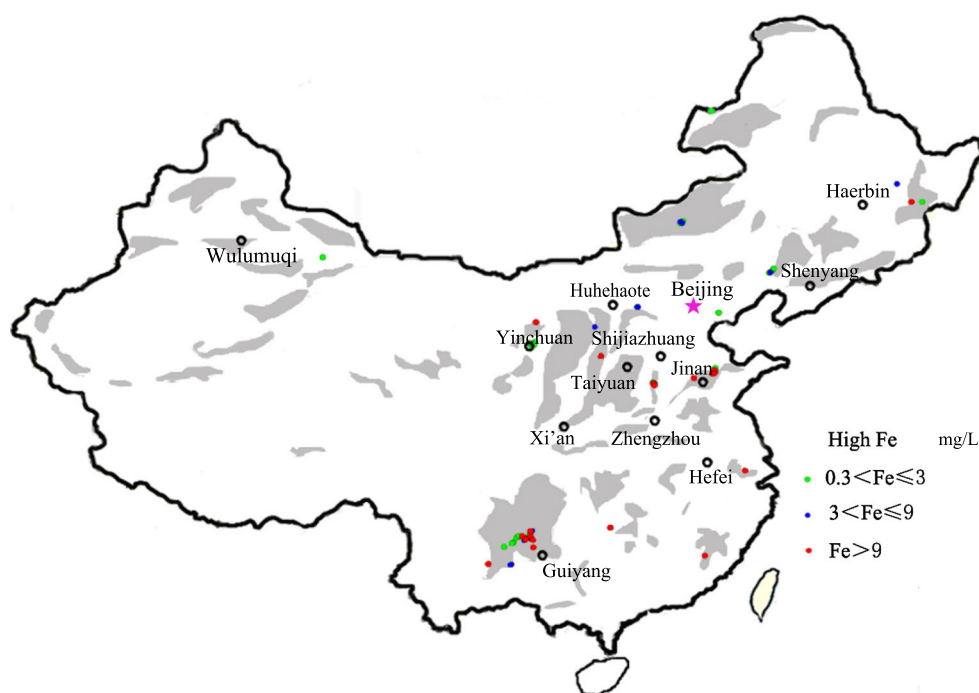


Fig. 6 Distribution of CMD with elevated iron in China



alkaline minerals, increasing Ca^{2+} , Mg^{2+} , and SO_4^{2-} concentrations. Also, in some mining areas, salt water intrusion can cause highly saline CMD. For example, the CMD in the Longkou coalfield of southwest Shandong region, which is near the sea, has high levels of Cl^- and relatively low SO_4^{2-} concentrations because of seawater intrusion. The Cl^- concentration for Wali, Beizao, and Liangjia mines in the Longkou coalfield is 930, 3,207, and 2,827 mg/L, respectively, while the respective SO_4^{2-} concentrations are 74, 81, and 88 mg/L, which is much less than the SO_4^{2-} concentrations of the other coalfields in the region.

It should be noted that even in the same coal mine, differences in exploitation location and depth can lead to differences in salinity. Generally, salinity increases with depth. For example, in the Zhaolou mine of the southwest Shandong region, the exploitation depth increased from 600 to 1,000 m during 2002–2009, and the salinity concentrations increased from 4,314 to 5,266 mg/L.

CMD with High Sulfate

Sulfate concentrations in the analyzed CMD samples fall in the range of 1–4,654 mg/L with a median of 328 mg/L (Table 1), which exceeds the groundwater quality standard. Non-acidic CMD with high sulfate is mostly located in southwest Shandong, Huainan-Huai-bei, Shendong, Jizhong, Ningdong, and Xinjiang (Fig. 4). There are two main causes of this CMD: sulfate produced by pyrite oxidation (followed by natural neutralization) and by dissolution of sulfur-bearing and gypsum minerals. For example, the Ordovician limestone aquifer, which is mostly composed of dolomite and is the main source of CMD in southwest Shandong, Huainan-Huai-bei, Jizhong, and other regions, often contains anhydrite.

CMD with High Fluoride

Fluoride concentrations greater than 1.0 mg/L is mainly found in northern China CMD, including the Huainan-Huai-bei, southwest Shandong, Henan, and Shanxi regions (Fig. 5). The CMD of the Panzhuang Mine of the Shanxi region has especially high F⁻ concentrations, up to 5.9 mg/L, far exceeding the groundwater standard. The coal-bearing stratum in these areas contains a small amount of fluorine-bearing minerals, such as fluorapatite ($\text{Ca}_5\text{F}(\text{PO}_4)_3$), cryolite (Na_3AlF_6), and fluorite (CaF_2). In addition, the strata contain magmatic intrusions that contain fluorine-bearing minerals that have dissolved into the groundwater after many years of physical and chemical reactions. Fluorine-bearing minerals and adsorbed fluorine in the soil are also F⁻ sources.

CMD with Elevated Iron and Manganese

In this study, CMD with Fe concentrations greater than 0.3 mg/L and Mn concentrations greater than 0.1 mg/L were considered high, based on Chinese groundwater quality standards. This type of CMD is mainly concentrated in the Yunnan-Guizhou region. In addition, CMD in Feicheng of southwest Shandong, Xishan of Shanxi, Hebi of Henan, Shizuishan of Ningdong, Jinzhushan of Hunan, and Yong'an of Fujian, also exceed these standards (Figs. 6, 7).

There are two reasons for high Fe and Mn in CMD. One is that iron- and manganese-bearing minerals can dissolve in groundwater; the other is FeS_2 oxidation. The acidity created by pyrite oxidation can react with manganese-bearing rock, increasing Mn levels. Most of the CMD in this study that had elevated Mn in this research had a pH of 6–9 (Fig. 8). When the pH was less than 6, Fe concentrations increased as pH decreased.

CMD Utilization and Treatment in China

Similar to the Mineral Council of Australia's Water Accounting Framework definition of Reuse Efficiency and Recycling Efficiency, the Chinese Ministry of Land and Resource has defined Utilization Efficiency of Coal Mine Water as the difference between generated CMD flow and discharged CMD flow to the environment as a proportion of generated CMD flow.

Utilization efficiency

$$= \frac{\text{Generated CMD flow} - \text{Discharged CMD flow}}{\text{Generated CMD flow}}$$

China's government is paying increased attention to the utilization and management of CMD each year. According to China's National Energy Administration, about 6.1 billion m^3 of CMD was generated in 2010 and the utilized CMD was about 3.6 billion m^3 , with an efficiency of 59 %, which was 28 % better than in 2005. The Mine Water Utilization Development Planning of China has stated that by the end of 2015, the national annual CMD discharge may reach 7.1 billion m^3 and that in order to increase utilization efficiency to 75 %, 5.4 billion m^3 of this will have to be used (NDRC 2012).

The Mine Water Utilization Development Planning of China has indicated that by the end of 2015, laws, regulations, macro-management, and technical support of mine drainage utilization should gradually be established to industrialize mine drainage utilization. By 2016, 448 projects, with a total investment of RMB 1.04 billion (over \$171 million U.S.), will be constructed to enhance CMD treatment and utilization capacity by 2.25 billion m^3 per year.

Fig. 7 Distribution of CMD with elevated manganese in China

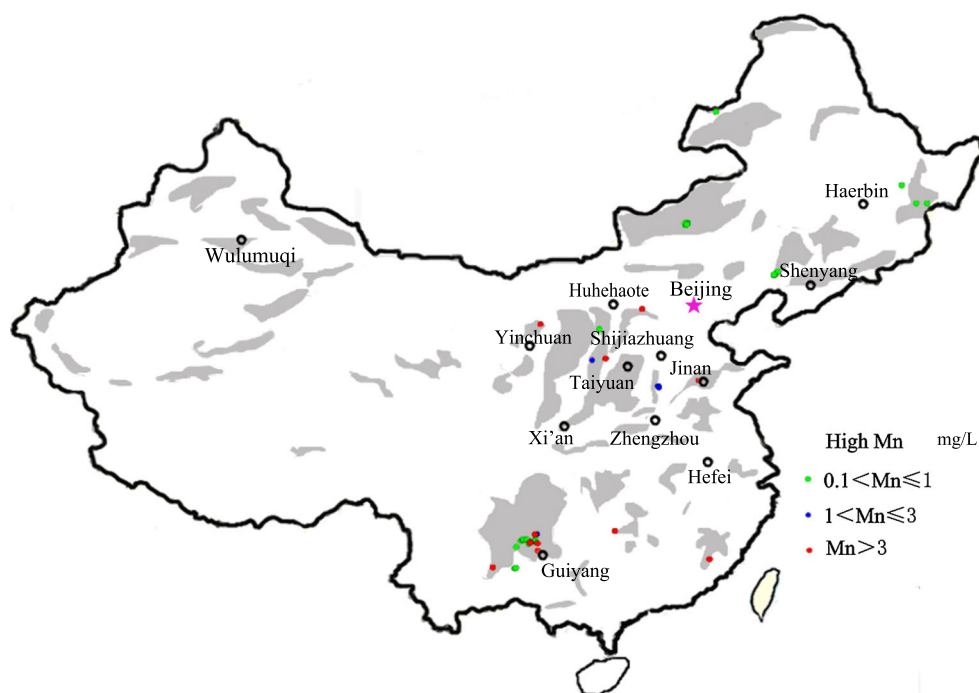
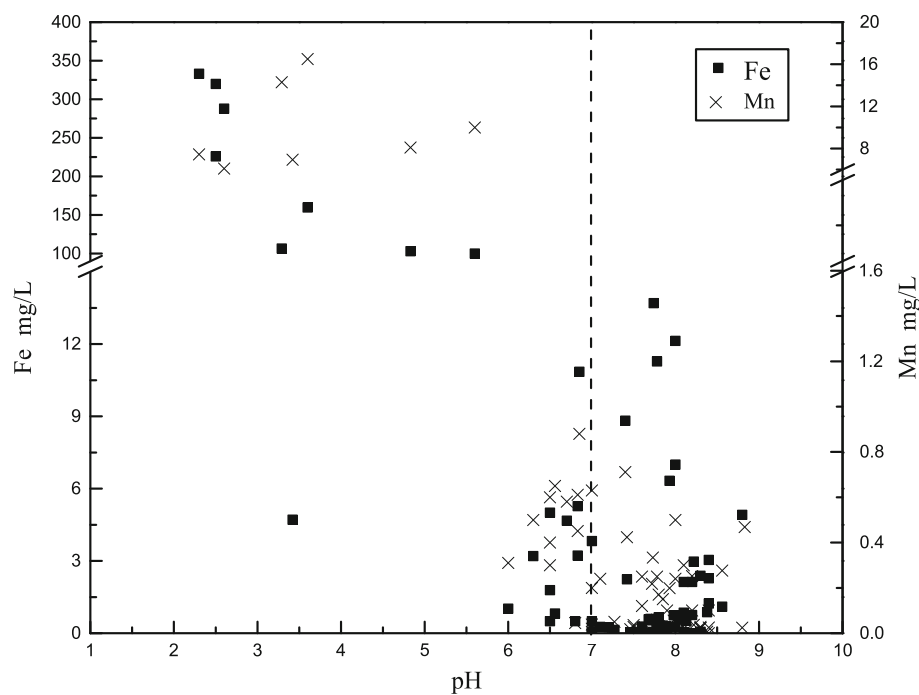


Fig. 8 The distribution of iron and manganese concentrations relative to pH in China's CMD



CMD utilization in China usually follows one of three approaches (Guo et al. 2008). Two only require simple treatment (i.e. flocculation, sedimentation, and filtration) to supply water for industrial use, which is less demanding in terms of water quality. After simple treatment, CMD can be used for coal production, coal washing, etc., which is presently 70 % of the total utilized CMD; it can also be used for planting and irrigation (currently accounting for

15 % of the utilized CMD). The third approach is through advanced treatment such as ion exchange, absorption, and reverse osmosis to supply water to residents of mining areas; this currently accounts for 10 % of the utilized CMD. In some arid areas of northern China, this treated CMD is the major water resource. Table 4 shows common treatment technologies for the different types of CMD (Chen et al. 2007; Cui et al. 2010; Guo et al. 2008; He et al.

Table 4 Typical treatment technologies for CMD in China

| CMD | Methods |
|----------------|---|
| Common | Sedimentation, flocculation, filtration |
| Acidity | Neutralization, microorganism, wetland, fly ash |
| High salinity | Ion exchange, membrane separation (electrodialysis, reverse osmosis), evaporation concentrates, diluted excretion |
| High sulfate | Lime, adsorption, ion exchange, flocculation |
| High fluorine | Chemical deposition, coagulation and sedimentation, active alumina, absorption |
| High Fe and Mn | Aeration and neutralization, oxidation, media filtration |

2010; Jiao 2012; Wang and Cheng 2007; Yang et al. 2009) Table 3.

Pollution prevention and management can effectively reduce CMD discharge and treatment costs. Pollution prevention and management is gradually being promoted and some success, such as pre-dewatering and limiting contact of groundwater with mine operations, has been achieved. For example, after limiting contact of groundwater with mine operations, 20,000–30,000 m³ a day of clean groundwater is being pumped from the Xinhe coal mine to supply the city of Xuzhou.

Conclusions

Over 6.1 billion m³ of CMD is being discharged annually in China, which is not only a serious environmental pollution problem, but also a waste of available resources, since most of China's coal-producing regions are in water-deficient areas and could benefit from CMD utilization. We analyzed the chemical characteristics and distribution of CMD based on analysis of discharge water from 269 coal mines in 11 large coal-producing regions in China. Six broad types of CMD were identified: slightly contaminated, acidic, high salinity, high sulfate, high fluoride, and CMD with elevated iron and manganese.

Acidic CMD is mainly located in southern China, where the lowest pH observed was 2, while little acidic CMD is located in northern China. Acidic CMD is often treated by chemical neutralization or passive treatment. In western China, the CMD often contains high levels of salinity, with 22,225 mg/L being the highest TDS detected. Saline CMD can be treated by ion exchange, reverse osmosis, or electrodialysis in order to meet drinking water quality standards. In eastern China, the salinity of CMD is generally less than 2,000 mg/L when shallow coal seams are being exploited, but rises as exploitation goes deeper. Flocculation and sedimentation are used for medium and low

salinity CMD to allow its use for coal production or agricultural irrigation. High sulfate CMD areas are mainly concentrated in southwest Shandong, Huainan-Huaiabei, Shanxi, Henan, and Ningdong. Lime, adsorption, and ion exchange are often used to treat this type of CMD.

CMD with high fluoride is mainly located in northern China, with the highest fluoride concentration detected being 5.9 mg/L; fluoride can be removed from CMD by active alumina, ion exchange, and absorption.

CMD with elevated iron and manganese is concentrated in the Yunnan-Guizhou region, while some occurs in other regions. This type of CMD can be treated by flocculation, sedimentation, absorption, ion exchange, and membrane separation.

In recent years, with the increase of environmental consciousness and water shortages, mine drainage treatment technologies have obtained considerable development and wide use in China. But the utilization efficiency of mine drainage is still only 59 %, so there is extensive room for enhanced CMD treatment and utilization. For China, it is important to improve relevant laws and regulations, strengthen research, develop better CMD treatment technologies, and encourage the utilization of CMD by economic and administrative means to protect the environment and relieve the water shortage of China's mining regions.

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