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Chemical characteristics of coal mine drainage and its impact on the environment in Shandong province, China

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ABSTRACT

In order to provide basis for the pollution control and comprehensive utilization of coal mine drainage (CMD), chemical characteristics and environmental impact of CMD in Shandong province were studied. In this paper, mine water quality of 65 mines distributed in 9 major coalfields in Shandong province were analyzed, and the results of which show that CMD can be summarized into the following types: common CMD, acid CMD,CMD with high total dissolved solid (TDS), CMD with high fluoride, and CMD with high sulfate.Chemical characteristics of CMD tend to be different depending on the hydro-geological conditions of mining areas. The discharge of CMD resulted in contamination on the water resource and the pollution characteristics are closely related to pollutants within the CMD.

Keywords: Mine water, Mine drainage, Chemical characteristics, Water contamination

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INTRODUCTION

Shandong province is located in the East China, faces the Bohai sea and the Yellow sea on the east, with total area of 1.57×10^5 km^2 and it is one of the 14 large coal-producing regions that had been assigned as focus of development by the 12th five-year plan along with 9 coalfields including Yanzhou, Jining, Xinwen, Zaozhuang, Longkou, Zibo, Feicheng, Juye, and Huanghebei (the Huanghebei is a planned coalfield without any producing mine).

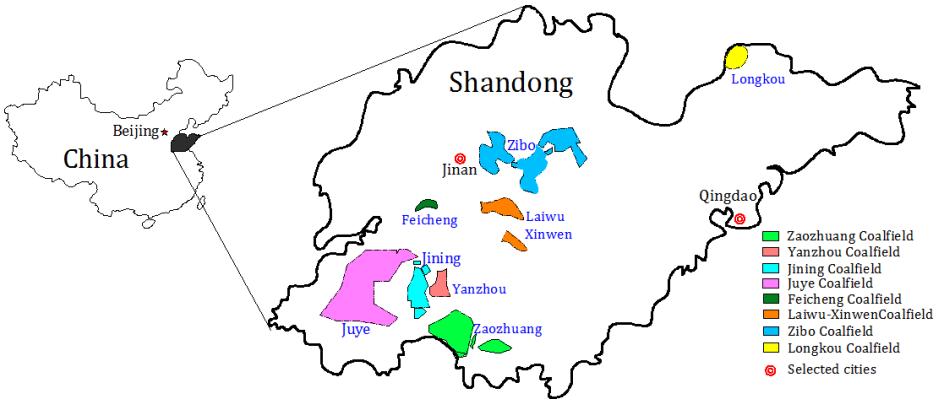

Figure 1: The location and distribution of major coalfields in Shandong province, China

At the moment there are in total 245 mines that are used for production in Shandong province and during 2004 to 2012 the annual coal production has maintained an average between 1.4-1.5 billion tons which is fifth place in the

country. It is predicted that the annual coal production in Shandong province will be stable at 1.5 billion tons in next ten years. Because of the limitation of complex geologic condition, coal production has become increasingly difficult and the average mining depth exceeds 600m with the maximum depth reaching 1350m.

CMD is generated by groundwater drainage during the mining process, and is mainly contaminated by coal dust and rock dust. Its characteristics are different from other industrialwastewater [1-3]. CMD can contain a variety of contaminants including organic, inorganic and bacteriological pollutants, poisonous and radioactive substances. Every year over 0.5 billion m^3 of CMD is discharged in Shandong province [4]. Part of CMD has been discharged into river or other surface water without treatment which caused water resource pollution. The reuse of CMD and water resource protection has become a key issue for coal industry development in Shandong province. In this paper, chemical characteristics and environmental impact of CMD in whole Shandong province-wide has been investigated and studied, which can provide basis for government to formulate policies and regulations related to mine water treatment and reuse.

EXPERIMENTAL SECTION

Chemical characteristics of CMD in Shandong province

In china CMD is normally classified into common CMD, CMD with high TDS, acid CMD as well as poisonous CMD [5, 6] depending on chemical characteristics and its feasibility as drinking water. Due to the fact that fluoride is the only regular poisonous substance that can be found in CMD within Shandong province, so in this investigation poisonous CMD is specifically named CMD with high fluoride. In addition, due to sulfate concentration of most CMD are high within Shandong province and the fact that sulfate has heavy impact on environment and CMD treatment, CMD with high sulfate is added to this investigation. In this paper a total of nine main coalfields in Shandong province including 65 typical mines are investigated. CMD quality statistical results are shown in Table 1.

Table 1: CMD quality of main coalfields in Shandong province, China (mg/L, not for pH)

Coalfields	Involved mines	pH (sample quantity)	F (sample quantity)	Fe (sample quantity)	TDS (sample quantity)	SO_4 (sample quantity)	References
Zaozhuang	10	4.20-8.40 (10)		57 (1)	569-3373 (10)	355-1907 (6)	[7, 8]
Yanzhou	6	7.00-8.75 (6)	0.91-1.40 (2)		290-2660 (6)	575-1380 (2)	[9]
Xinwen- Laiwu	11	7.05-8.23 (11)	2.92 (1)		441-1759 (11)	302-1095 (5)	[10]
Jining	4	6.00-9.00 (3)			1050-1749 (3)	445 (1)	
Longkou	3	7.80-8.35 (3)	0.37-0.62 (3)		2366-5522 3)		[11, 12]
Feicheng	9	3.22-9.00 (9)		334-691 (2)	403-2569 (9)	1806 (1)	[13]
Zibo	16	2.71-8.10 (16)	0.19-0.65 (3)	0-478 (15)	579-4331 (14)	113-3410 (16)	[14-16]
Juye	6	7.26-7.86 (6)	0.87-2.18 (6)		2472-5266 (6)	1301-3160 (6)	

(Part of data was collected from publications)

Common CMD

This type of CMD is widely distributed in all coalfields in Shandong province, accounting for 32.8% of all CMD samples, usually contains pollutants such as suspended solid (SS), chemical oxygen demand (COD), oil, and lower concentration of organic contaminants. The characteristics of this type of CMD are high SS, no special pollutants, neutral pH, and grey or black color. SS comes mainly from coal dust and rock dust which are produced in mining process. COD, oil and organic pollutants come from waste oil of mining equipments, human excrement and other anthropogenic emissions.

CMD with high TDS

This type usually refers to CMD with TDS concentration higher than 1000 mg/L and is widely distributed in all coalfields of Shandong (Figure 2). According to statistical results of Table 1, CMD in Shandong has TDS between 290 to 5266 mg/L with an average of 1801 mg/L, among whichTDS in the range of 1000-2000 mg/L accounted for 28.3%, TDS in the range of 2000-4000 mg/L accounted for 28.3%, and TDS greater than 4000 mg/L accounted for 8.3%.In coalfields such as Zibo, Juye and Longkou, the TDS of CMD can get higher than 4000 mg/L. Especially in Zhaolou mine of Juye coalfield, the maximum TDS being up to 5266 mg/L.

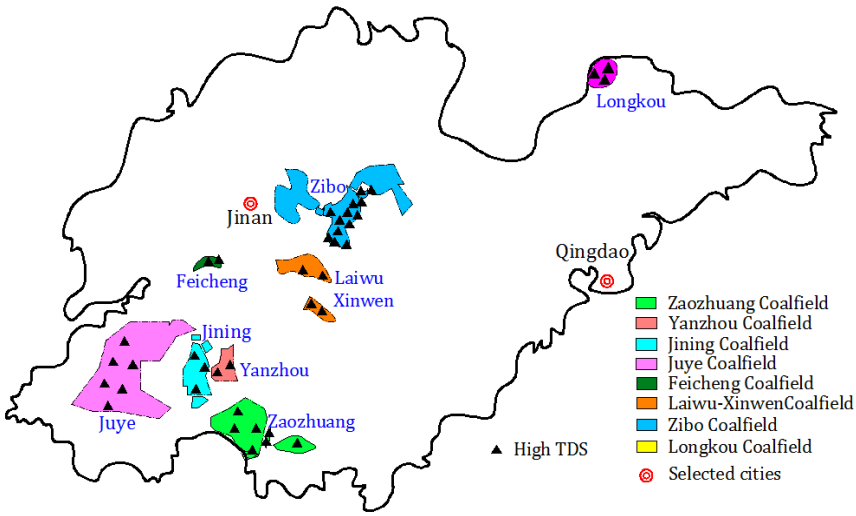

Figure 2: The distribution of CMD with high TDS in Shandong

CMD with high TDS in these areas of Shandong province is formed due to the following reasons:

I) Coal measurestrata in these areas contains large amounts of carbonates and sulfate rocks, which accelerates dissolution of soluble mineral due to extensive contact with the groundwater during the mining process, as a result increasing the amount of salinity.

II) Low precipitation and high evaporation leads to groundwater concentration and results in higher groundwater salinity.

III) There is generally high sulfur in coal seams of these areas. During the exploitation of these coal seams, free acid, which is produced due to sulfide oxidation, reacts with carbonate minerals and alkaline minerals resulting the increasing of Ca^{2+}, Mg^{2+} and . The chemical equations are as follows:

$$4FeS_2 + 14H_2O + 15O_2 = 4Fe(OH)_3 + 8\uparrow SO_4^{2-} + 16H^+ \quad (1)$$

$$CaCO_3 + 4H^+ = Ca_2^+ + 2H_2O + CO_2\uparrow \quad (2)$$

$$MgCa(CO_3)_2 + 4H^+ = Mg^{2+} + Ca^{2+} + 2H_2O + 2CO_2\uparrow \quad (3)$$

IV) As a coalfield near sea, CMD with high TDS in Longkou coalfield has different chemical characteristics such as higher Cl^- concentration and lower concentration because of seawater intrusion. In this investigation the Cl^- concentration for Wali , Beizao, and Liangjia mine within longkou coalfield is 930ml/L, 3207ml/L and 2827ml/L respectively at the same time the concentration respectively is 74ml/L, 81ml/L and 88ml/L which is greatly below concentration of other coalfields within Shandong province.

It should be noted that even in the same coal mine, differences in exploitation location and depth leads to differences in hydrodynamic conditions, which in turn affects the TDS of CMD. Generally, TDS significantly increased with exploitation depth getting deeper in Shandong province. For example, in Zhaolou mine of Juye coalfield, the exploitation depth increased from 600m to 1000m during 2002-2009, among which the TDS of CMD increased from 4314 mg/L to 5266 mg/L.

Acid CMD

This type of CMD has pH value below 6. As shown in Figure 3, acid CMD is distributed in three coalfields located in Zibo, Feicheng and Zaozhuang. Especially, the CMD in Xihe mine of Zibo coalfield has pH value down to 2.71.

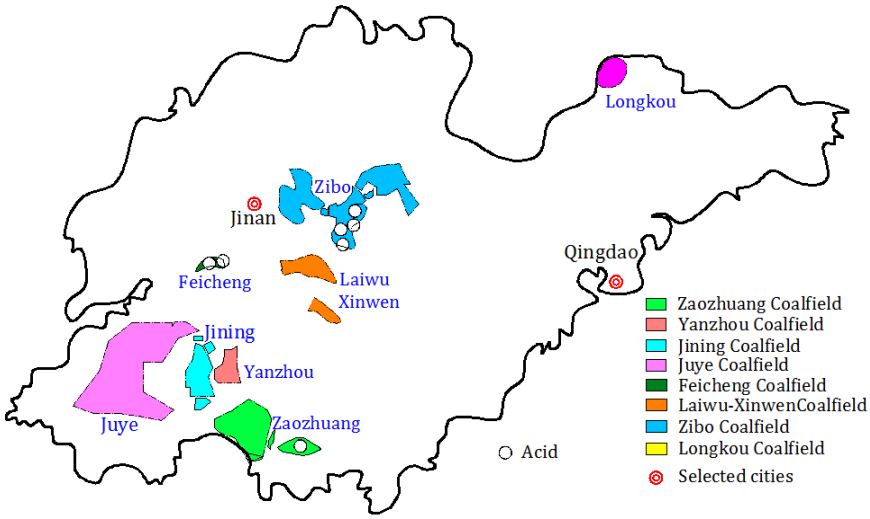

Figure 3: The distribution of acid CMD in Shandong

In Shandong province, the formation of acid CMD is generally due to the varying amounts of sulfur with mass fraction between 0.3% - 5.0% found in the coal seam mainly in the form of pyrite (FeS_2). These sulfur content ultimately forms sulfuric acid and iron ions through oxidation of micro-organisms and oxygen turning certain mine water acidic [17-19]. The chemical equations are as follows:

$$2FeS_2 + 2H_2O + 7O_2 = 2Fe^{2+} + 4SO_4^{2-} + 4H^+ \quad (4)$$

$$12FeSO_4 + 6H_2O + 3O_2 = 8Fe^{3+} + 12SO_4^{2-} + 4Fe(OH)_3 \quad (5)$$

$$2Fe^{3+} + 3SO_4^{2-} + 6H_2O = 2Fe(OH)_3 + 6H^+ + 3SO_4^{2-} \quad (6)$$

One of the characteristics of acid CMD is high iron ion concentration. In this investigation there are 7 mines that produce acidic CMD and iron ion concentration at the same time exceeds standard of 0.3 mg/L according to the Chinese Groundwater Quality Standard (GB-T14848-93).

CMD with high fluoride

This type generally refers to CMD with fluoride concentration more than 1.0 mg/L. During this investigation there are 7 mines that has CMD which can be classified as high fluoride CMD and 5 amoung which is located within Juye coalfield (Figure 4). Juye region has relatively high geochemical background value in terms of fluoride. The rocks in this region contain fluoride-rich minerals such as fluorapatite ($Ca_5F(PO_4)_3$), cryolite (Na_3AlF_6) and fluorite (CaF_2). Fluoride composition of groundwater is produced by these fluoride-rich minerals after long years of physical and chemical reactions [20].

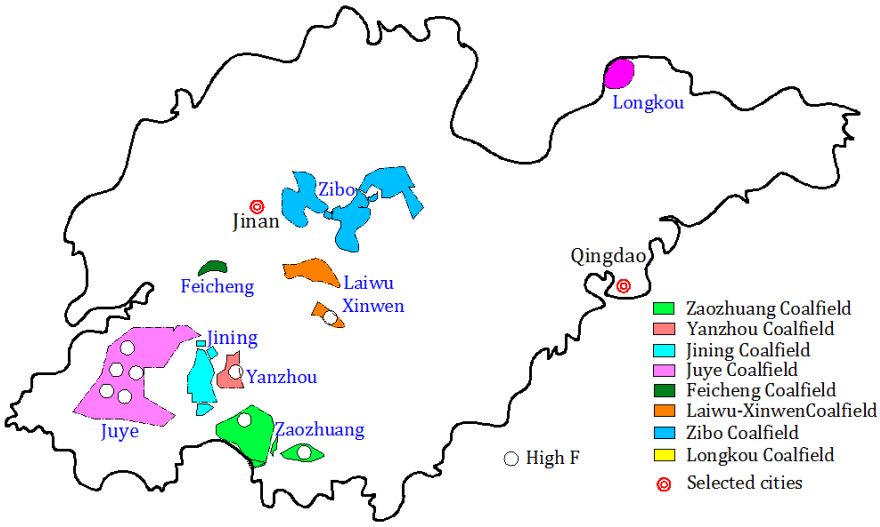

Figure 4: The distribution of CMD with high fluoride in Shandong

CMD with high sulfate

This type usually refers to CMD with sulfate concentration more than 250.0 mg/L, and it is widely distributed in all

coalfields apart from Longkou (Figure 5).

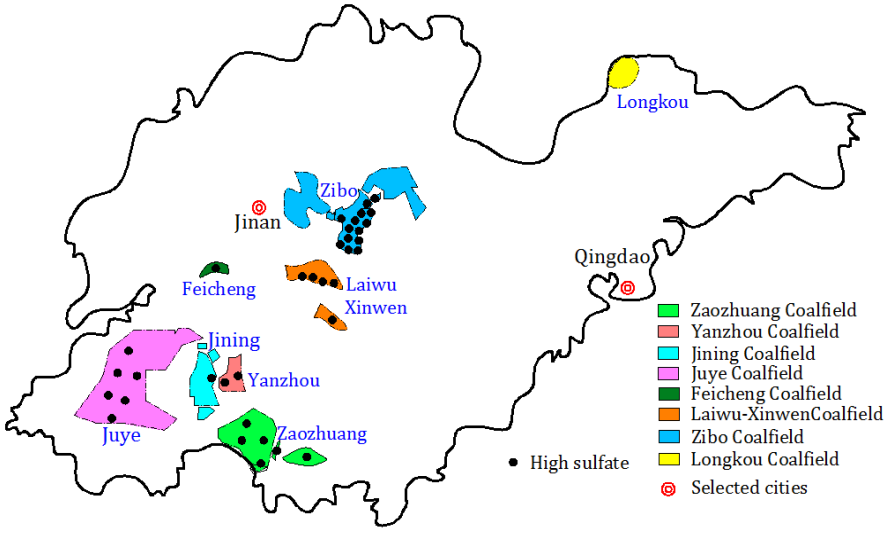

Figure 5: The distribution of CMD with high sulfate in Shandong

According to statistical results of Table 1, sulfate concentration of CMD falls in the range of 74 to 3410 mg/L with mean of 1267 mg/L, 4 times higher than the standard value specified by the Chinese Groundwater Quality Standard (GB-T14848-93). CMD with high sulfate is one branch of CMD with high TDS, and in this investigation, apart from Longkou coalfield all high TDS CMD found are at the same time high sulfate CMD, in addition the causes for both are the same. Moreover, the groundwater chemistry type changes from bicarbonate type to sulfate type after the groundwater salinity increasing because of the exploitation depth goes deeper, is also the reason for increase in sulfate concentration.

Environmental impact of CMD in Shandong province

The discharge of CMD has great impact on the environment of Shandong Province. For example there are many coal mines in the Xiaofu river valley within Zibo coalfield and each year about 50 million m^3 of CMD is discharged into the Xiaofu River and its tributaries [21]. According to 6 inspections made between year 1979 and 1996 the sulfate concentration, the total hardness, and the TDS has made obvious increase with the greatest increase being 3.5 times, 2.2 times, 2.6 times the original amount respectively (Figure 6). Groundwater beside the stream may be contaminated through recharging by seepage of stream.

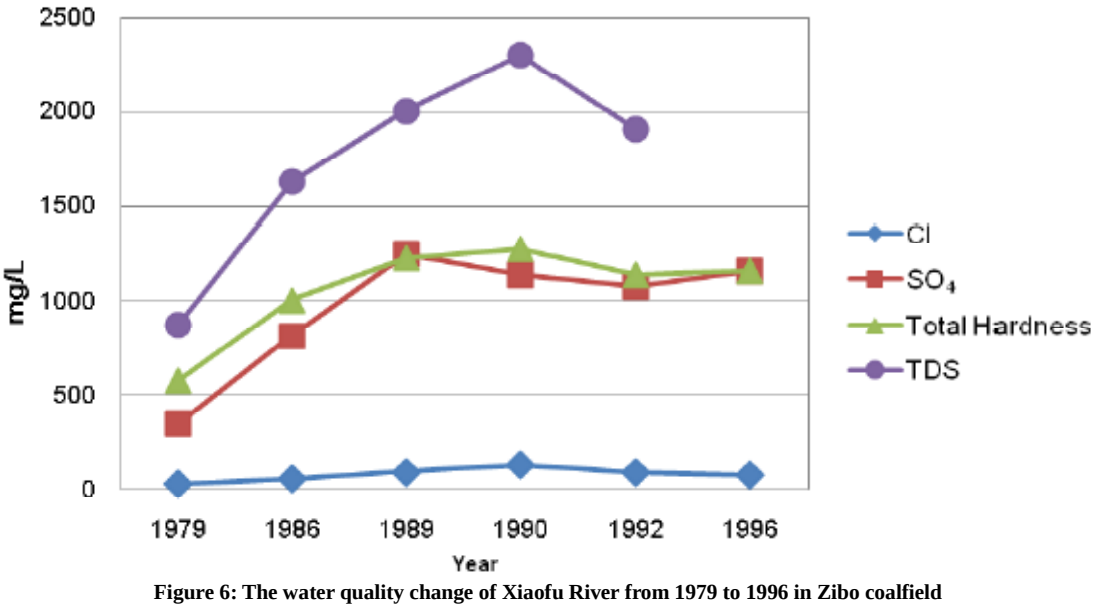

Figure 6: The water quality change of Xiaofu River from 1979 to 1996 in Zibo coalfield

Since water shortage is a serious problem in Shandong, CMD is directly used as irrigation without proper treatment in some mining areas and severer impact will be brought on agriculture, such as decreasing soil pH, soil hardening, and soil salinization.

CONCLUSION

Shandong province is one of the major coal-producing regions in China. Every year over 0.5 billion m^3 of CMD is discharged in Shandong province. The discharge of CMD resulted in contamination on the water resource and the pollution characteristics are closely related to pollutants within the CMD. Based on the CMD investigation and analysis results of 65mines distributed in 9 major coalfields in Shandong province, CMD can be summarized into the following types: common CMD, acid CMD, CMD with high total dissolved solid (TDS), CMD with high fluoride, and CMD with high sulfate. Chemical characteristics of CMD tend to be different depending on the hydro-geological conditions of mining areas.

With the rapid economic development, the growing water demand is causing pressing need for mine water treatment and reuse. Because of the complexity of CMD quality, appropriate treatment technologies and pollution prevention measures can be chosen according to the characteristics of coal mine water in order to protect water resource. Besides, this study can provide basis for government to formulate policies and regulations related to mine water treatment and reuse. For Shandong province, it is important to improve Environmental Impact Assessment regulation, strengthen environmental laws enforcement, and encourage the reuse of CMD by economic and administrative means with the goal to increase the reuse rate of CMD and relieve the water shortage in Shandong province.

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