Evaluation of treatment efficiency of coal mine drainage and impact on surrounding waters for selected coal mines

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Abstract. Coal mine water usually has high content of trace elements, leading to contamination of surrounding water bodies. Water treatment technologies are used to remove organic and inorganic matters from coal mine water. To evaluate the treating efficiency of conventional and advanced treatment on coal mine drainage, samples of coal mine, carbonate, surface water, and treated water from the treatment processes were collected and analysed. It turns out that the conventional treatment processes can hardly remove both major ions and trace elements in coal mine water. Reverse osmosis technology can reject major ions effectively, but weak in trace elements removal. The analysis of surface water quality suggested contamination by coal mine water in the coal-mine district. To control the contamination and reuse the coal mine water effectively and safe, it is necessary to evaluate more carefully the efficiency of water treatment and potential environment impact of coal mine water on the surrounding surface and ground waters.

1. Introduction
China has a fast-developing market, requiring huge coal assumption. At the same time, coal mine water is produced accompanying the mining activity. Up to 2018, coal mine water drainage has nearly 8 billion m³, accounting for more than 80% of all types of mining water, some of them are seemed as resource to reused for living and industry purpose. According to the content, the coal mine water can be divided into several types: clean, turbid, high mineralized, acid/alkaline, high [Fe]/[Mn], high radioactive content, high [F], and high trace element content water. The coal mine water usually has characteristics of high TDS, high hardness, and high turbidity, some of them are acid water and enriched in trace elements. The trace elements water is usually formed accompanying the acid coal mine water and has high contamination potential of surrounding surface and ground waters. To improve the water quality, the water needs to be treated strictly before drinking, irrigating, and other form of service.

In order to remove contaminates in the coal mine water, it is treated by various conventional and advanced technologies, including coagulation-flocculation, filtration, biologically active carbon, reverse osmosis, etc. The technology of coagulation-flocculation is a traditional method and widely used. By adding iron or aluminum ions and mix rapidly in a chamber, colloidal species and small particles in the water are aggregated into larger, heavier clusters that settle out in a short time. Reports of the technology used in the drinking water treatment are intensively. However, fewer researches could be found on the coal mine water treatment, especially on the removal of trace elements. For the oilfield produced water, the suspended and colloidal oil can be significantly removed [1-2], but not
effective for removing dissolved constituents unless they precipitate at the treatment conditions of the coagulation process. Some improved and enhanced coagulation-flocculation method have been developed, such as inorganic mixed metal (Fe, Me, and Al), ozone-enhanced coagulation [3], polynuclear polymer [4], and electrocoagulation [5]. In the filtration process, finite-size particles can be removed. While using granular filtration, particles are filtered by the effect of electrostatic interactions between these particles and filter media [6]. In the depth filtration, the colliding adheres to the filter media, and the particles that much smaller than the smallest pore size can be captured. Some adsorbents are also used in the adsorption process, such as zeolite, activated carbon, natural organic materials, organoclay, and synthetic polymers [7-8]. Comparing with the mentioned approaches, available technologies to remove trace elements in coal mine water includes reverse osmosis and adsorption. Shortages of RO includes high cost, restricted osmotic pressure and membrane fouling. Usually, the RO treatment is used after coagulation-flocculation, and filtration.

Chinese government has promulgated laws and regulations to constrain mining and discharge activities to improve quality of coal mine water. The Chinese standard of disposing by coal mine industry (GB/T14848-2017) regulated 11 items, including trace elements of Hg, Cd, Cr, Pb, As, F, etc. The transform administrative fees to taxes for the coal mine water discharging could also push the owners and managers of coal mines to elevate level of coal mine water treatment and reuse. Although some of the coal mine water has been treated before discharged or reuse, the quality may not satisfy. At the same time, part of the coal mine drainage is directly disposed. Therefore, the water bodies in the coal mine district may be impacted, which threat human health. This paper aims to evaluate the quality of coal mine water, potential impact on surrounding water, in a selected district, and treatment effect of conventional and RO technology on removal of major ions and trace elements. Coal mine, carbonate, surface water, and treated water from the treatment processes were collected and analyzed. The results of coal mine water and treated water are used to evaluate the treating efficiency of major ions and trace elements. The samples of surface water, combining the coal mine water, is used to evaluate impact of coal mine water on surrounding water bodies.

2. Site description and method

2.1. Site description

Xuzhou-Datun coal mine district was chosen as the study area, including Sanhejian coal mine of Xuzhou coal mining group, and Kongzhuang, Longdong, Yaoqiao coal mines, of Datun Coal and Electricity Group, which is shown in the Figure 1. The study area is located in the northwest of Jiangsu province, northern China, with average temperature, relative humidity and air pressure, 14 °C, 73%, 101280 Pa, respectively. Average precipitation is 758 mm, ranging from 492 mm to 1178 mm, and average evaporation is 1623 mm. A series of sediment stratum Simian, Cambrian, middle-lower Ordovician, middle-upper Carboniferous, Permian, Jurassic, Cretaceous, Tertiary and Quaternary system, cover the Archean system.

The active mining seams are in the Carboniferous and Permian systems, the former includes Benxi and Taiyuan formations, and the latter includes Shanxi formation (coal seam No. 7&9) and Lower-Shihezi formations (coal seam No. 2), listed from bottom to top in both systems. In the lower formation, white feldspar, quartz granule-sandstone and silicon-mudstone cementation are the main minerals. Grey mudstone, sand-mudstone and sandstone are the main rocks in the middle Shanxi formation with some silicon-mudstone and siderite also present [9]. In the upper formation, grey-green middle-fine quartz sandstone, siltstone, mudstone are the main minerals.

2.2. Applied water treatment

The coal mines of Datun Coal and Electricity Group [10-11] and Xuzhou coal mining group [12-13] have applied water treatment system to treat the coal mine water reusable. Two type of systems are used based on application of water, basic treatment systems and advanced treatment systems.
In the basic systems, it is composited by pre-sedimentation, coagulation and flocculation, multimedia filtration, disinfection is used in some coal mines. In an advanced treatment system, the active-carbon filtration, advanced filtration, electro-dialytic treatment and reverse osmosis treatment are used. The water after advanced treatment satisfies standard of industry water discharging in some indexes, including pH, COD, BOD, suspended solid, and NH$_3$-N. Then the water is discharged directly, or used in coal washing, civil and industrial application. It is not mentioned how much of the trace elements be removed.

2.3. Method
Twenty-one water samples were collected, including ten coal mine water, six treated water by conventional method, one RO treated water samples, two carbonate and two surface water samples. The coal mine water samples were collected from Xuzhuang, Longdong, Sanhejian, and Zhangshuanglou coal mines. Carbonate water collected from limestone seam underground from the observing hole in the Sanhejian coal mine. The surface water samples were collected from coal mine district and non-coal mine district, respectively. Treated water samples were collected from the water treatment system, including the water in setting tank, overflow and bottom flow from the thickener, coal-washing water, and post-RO water. 1000 mL Nalgene bottles were used to contain water samples, which were cleaned by acid in laboratory and rinsed twice before the sample were collected.

Major ions and physical parameters of water samples were determined in Jiangsu Provincial Coal Geology Research Institute in line with Chinese standard protocols. Concentration of trace elements water samples were determined by ICP-AES. K and Na were analyzed by flame atomic absorption spectrophotometry (GB 11904-89). Mg and Ca were measured using atomic absorption spectrophotometric (GB 11905-89). Total dissolved solids and hardness were analyzed in term of the standard GB/T 8538 method. Fe, Sulphate and chloride were determined using flame atomic absorption spectrophotometry (GB 13196-91) and silver nitrate titration (GB 11896-89), respectively.

The ICP-AES analysis was carried out in the Nanjing University using a JY38S ICP-AES model. Limit of detection and deviation for the analysis were 0.01 µg/mL and less than 2 %, respectively.

3. Trace element removal and concentrations in surrounding waters

3.1. Removal efficiency of major ions and trace elements during treatment
Table 1 shows major and trace elements in 21 water samples. The figure 2-4 show boxplots of major ions and some trace element concentrations in different type of water samples, representing the coal mine water, groundwater, surface water, conventional treated and RO treated coal mine water samples. Although the conventional treated technologies include flocculation, filtration, detention, etc, there have no significant difference to divide them. Therefore, the concentration data during the conventional treated process were calculated as a group. The table and figures are used to compare the concentrations in drainage, conventional treated and reverse osmosis membrane treated and evaluate the potential impact of coal mine water on surrounding waters.
### Table 1. Major ion concentrations, parameters, and trace elements in water samples.

| No. | Water type | 
|-----|------------|
|     |            | mg/L \(\pm\) |
| 1   | Total mine water | 13.7 ± 0.1 |
| 2   | Coal mine water | 1.4 ± 0.6 |
| 3   | Coal mine water | 26.0 ± 1.2 |
| 4   | Coal mine water | 400.8 ± 26.0 |
| 5   | Coal mine water | 1402.0 ± 140.2 |
| 6   | Coal mine water | 263.8 ± 16.0 |
| 7   | Coal mine water | 321.0 ± 18.0 |
| 8   | Coal mine water | 1151.3 ± 140.2 |
| 9   | Coal mine water | 192.0 ± 12.0 |
| 10  | Coal mine water | 60.0 ± 4.0 |
| 11  | Coal mine water | 23.5 ± 1.0 |
| 12  | Coal mine water | 400.0 ± 24.0 |
| 13  | Coal mine water | 31.7 ± 1.0 |
| 14  | Coal mine water | 6.4 ± 0.1 |
| 15  | Coal mine water | 14.9 ± 0.7 |
| 16  | Coal mine water | 6.1 ± 0.3 |
| 17  | Coal mine water | 10.7 ± 0.5 |
| 18  | Coal mine water | 20.9 ± 1.0 |
| 19  | Coal mine water | 20.9 ± 1.0 |
| 20  | Coal mine water | 0.4 ± 0.0 |
| 21  | Coal mine water | 1.0 ± 0.0 |
| 22  | Coal mine water | 3.0 ± 0.1 |
| 23  | Coal mine water | 0.0 ± 0.0 |
| 24  | Coal mine water | 0.0 ± 0.0 |
| 25  | Coal mine water | 0.0 ± 0.0 |
| 26  | Coal mine water | 0.0 ± 0.0 |
| 27  | Coal mine water | 0.0 ± 0.0 |
| 28  | Coal mine water | 0.0 ± 0.0 |
| 29  | Coal mine water | 0.0 ± 0.0 |
| 30  | Coal mine water | 0.0 ± 0.0 |
| 31  | Coal mine water | 0.0 ± 0.0 |
| 32  | Coal mine water | 0.0 ± 0.0 |
| 33  | Coal mine water | 0.0 ± 0.0 |
| 34  | Coal mine water | 0.0 ± 0.0 |
| 35  | Coal mine water | 0.0 ± 0.0 |
| 36  | Coal mine water | 0.0 ± 0.0 |
| 37  | Coal mine water | 0.0 ± 0.0 |
| 38  | Coal mine water | 0.0 ± 0.0 |
| 39  | Coal mine water | 0.0 ± 0.0 |
| 40  | Coal mine water | 0.0 ± 0.0 |
| 41  | Coal mine water | 0.0 ± 0.0 |
| 42  | Coal mine water | 0.0 ± 0.0 |
| 43  | Coal mine water | 0.0 ± 0.0 |
| 44  | Coal mine water | 0.0 ± 0.0 |
| 45  | Coal mine water | 0.0 ± 0.0 |
| 46  | Coal mine water | 0.0 ± 0.0 |
| 47  | Coal mine water | 0.0 ± 0.0 |
| 48  | Coal mine water | 0.0 ± 0.0 |
| 49  | Coal mine water | 0.0 ± 0.0 |
| 50  | Coal mine water | 0.0 ± 0.0 |
| 51  | Coal mine water | 0.0 ± 0.0 |
| 52  | Coal mine water | 0.0 ± 0.0 |
| 53  | Coal mine water | 0.0 ± 0.0 |
| 54  | Coal mine water | 0.0 ± 0.0 |
| 55  | Coal mine water | 0.0 ± 0.0 |
| 56  | Coal mine water | 0.0 ± 0.0 |
| 57  | Coal mine water | 0.0 ± 0.0 |
| 58  | Coal mine water | 0.0 ± 0.0 |
| 59  | Coal mine water | 0.0 ± 0.0 |
| 60  | Coal mine water | 0.0 ± 0.0 |
| 61  | Coal mine water | 0.0 ± 0.0 |
| 62  | Coal mine water | 0.0 ± 0.0 |
| 63  | Coal mine water | 0.0 ± 0.0 |
| 64  | Coal mine water | 0.0 ± 0.0 |
| 65  | Coal mine water | 0.0 ± 0.0 |
| 66  | Coal mine water | 0.0 ± 0.0 |
| 67  | Coal mine water | 0.0 ± 0.0 |
| 68  | Coal mine water | 0.0 ± 0.0 |
| 69  | Coal mine water | 0.0 ± 0.0 |
| 70  | Coal mine water | 0.0 ± 0.0 |
| 71  | Coal mine water | 0.0 ± 0.0 |
| 72  | Coal mine water | 0.0 ± 0.0 |
| 73  | Coal mine water | 0.0 ± 0.0 |
| 74  | Coal mine water | 0.0 ± 0.0 |
| 75  | Coal mine water | 0.0 ± 0.0 |
| 76  | Coal mine water | 0.0 ± 0.0 |
| 77  | Coal mine water | 0.0 ± 0.0 |
| 78  | Coal mine water | 0.0 ± 0.0 |
| 79  | Coal mine water | 0.0 ± 0.0 |
| 80  | Coal mine water | 0.0 ± 0.0 |
| 81  | Coal mine water | 0.0 ± 0.0 |
| 82  | Coal mine water | 0.0 ± 0.0 |
| 83  | Coal mine water | 0.0 ± 0.0 |
| 84  | Coal mine water | 0.0 ± 0.0 |
| 85  | Coal mine water | 0.0 ± 0.0 |
| 86  | Coal mine water | 0.0 ± 0.0 |
| 87  | Coal mine water | 0.0 ± 0.0 |
| 88  | Coal mine water | 0.0 ± 0.0 |
| 89  | Coal mine water | 0.0 ± 0.0 |
| 90  | Coal mine water | 0.0 ± 0.0 |
| 91  | Coal mine water | 0.0 ± 0.0 |
| 92  | Coal mine water | 0.0 ± 0.0 |
| 93  | Coal mine water | 0.0 ± 0.0 |
| 94  | Coal mine water | 0.0 ± 0.0 |
| 95  | Coal mine water | 0.0 ± 0.0 |
| 96  | Coal mine water | 0.0 ± 0.0 |
| 97  | Coal mine water | 0.0 ± 0.0 |
| 98  | Coal mine water | 0.0 ± 0.0 |
| 99  | Coal mine water | 0.0 ± 0.0 |
| 100 | Coal mine water | 0.0 ± 0.0 |

3.1.1 Major ions. The conventional treatment technologies have a significant effect on removing suspended solid, organic matter, odor, according to the treating system report. The total dissolved solid, hardness, and major elements, however, are less mentioned.

According to the test results, the median value of TDS in coal mine water and treated water were 3620 mg/L, 2140 mg/L, hardness values were 527.4 mg/L and 699.9 mg/L, respectively. It is shown that the TDS was partly removed, while increased [Ca] and [Mg] leaching to higher hardness values. Compared to the national standard, “Quality standard for ground water” regulate the TDS higher than 2000mg/L and hardness higher than 550 as water type V. The “Standards for drinking water quality” regulate highest values of TDS and hardness 1000 mg/L and 450mg/L, respectively.

For the specific elements/ions, Na + K, Ca, Mg, Cl, SO\(_4\)\(^{2-}\) and HCO\(_3\) were removed by 38.4%, -31.9%, -2.8%, 16.6%, 24.0%, and 5.1% based on median values. The national standards “Environmental quality standards for surface water” and “Standards for drinking water quality” regulate that [SO\(_4\)\(^{2-}\)] and [Cl] should lower than 250 mg/L in the water of domestic water system. The water both before and after conventional treatment has break this limit.

A much better removal efficiency of major ions was observed by the RO membrane treatment. After the treatment, the median values of TDS and hardness values were 40 mg/L and 39.79 mg/L. Major elements Na + K, Ca, Mg, Cl, SO\(_4\) and HCO\(_3\) were removed by 99.8%, 96.0%, 88.0%, 96.2%, 100%, 92.1%, respectively.
3.1.2. Trace elements. In the coal mine water, As, Hg, Se, Cd, Pb, Cr belonged to water type III/I, V/I, IV/V, III/II, III/III, and I/-, when compared median values with the “Quality standard for ground water” and “Environmental quality standards for surface water”. After the conventional and reverse osmosis treatment, water types regarding the trace elements didn't change. Compared with “Standards for drinking water quality”, the values of trace elements after RO treatment and the highest acceptable value in drinking water are 27.9 / 10 mg/ml for arsenic, 0.8 / 5 mg/ml for cadmium, 3.2 / 50 mg/ml for chromium, 37.1 / 10 mg/ml for lead, 5.4 / 1 mg/ml for mercury, 28 / 10 mg/ml for selenium, respectively. The results suggest that the RO treated water is not suitable for drinking regarding concentrations of trace elements.

The remove efficiency by conventional and RO membrane on trace elements were calculated, which are shown in the table 2.

| Water treatment technologies | As  | Hg  | Se  | Mo  | P   | Zn  | Cd  | Sb  | Pb  | Co  | Ni  | Ba  |
|-----------------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Conventional                | 2.3%| -50.0%| 8.1%| -0.2%| -79.4%| -16.0%| -22.2%| -16.7%| -41.4%| -9.8%| -25.0%| -53.1% |
| RO membrane                 | 23.7%| -20.0%| 37.7%| 93.8%| 9.2%| 25.9%| 40.7%| 6.1%| -26.0%| 9.8%| 17.4%| 80.6% |

| Water treatment technologies | B  | Si  | Mn  | Fe  | Cr  | V   | Ag  | Ti  | Sc  | Al  | Sr  |
|-----------------------------|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Conventional                | 15.8%| 1.1%| 36.6%| 81.2%| -6.8%| 28.3%| -7.4%| 3.9%| 20.0%| -77.2%| -12.3% |
| RO membrane                 | 71.0%| 55.0%| 80.2%| 96.8%| -8.5%| 97.2%| -124.8%| 78.6%| -40.0%| 86.0%| 99.0% |

The figure 3 and 4 are concentration boxplots for selected trace elements in coal mine water, conventional and RO treated coal mine water samples, carbonate water and surface water samples. As shown in the table 2, figure 3 and 4, trace elements showed different behaviors during the conventional and RO treatment process. After the conventional treatment, concentrations of Fe and Mn were significantly reduced in the treated water, while Hg, P, Ba, Al elevated.

After the RO treatment, 23.7 % of arsenic was removed by the RO technology. Ning [14] found the arsenic with oxidation states (V) was much more effectively rejected than arsenic (III), Akin et al [15] argued that Solution pH at 4.1 and 9.1 was required for removing of As (V) and As (III), respectively. At the same time, pressure is important on the arsenic rejection. In the studied plant, boron was removed by 15.8% using the conventional method, and by 71.0% by the RO technology, while Oo and Song [16] found that 99% of boron was removed at high pH and 40%-80% was removed at neutral pH, Yavuz et al [17] found 94.5 % - 95 % of boron was rejected at a high pH value of 10.5 and operating
pressure of 12 bar. Removal efficiency for cadmium was 40.7% in our study. Qdais and Moussa [18] reported that high removal efficiency (98% for copper and 99% for cadmium) can be achieved by the osmosis reverse membrane. Lin argued that the RO concentrate can effectively remove metal elements, such as lead and copper, as they form strong inner-sphere complexes between the metal cations and coagulation media- Fe/AL oxide surface, while selenate is a weakly bonding anion typically assumed to form outer-sphere surface complexes with Fe/Al oxides [19]. In our study, removal efficiency for some metal elements was high, while some others, such as Hg, Pb, Ag, was relatively low. The results suggest that the RO membrane differ in removing trace elements in coal mine water. The impact factors on the removal may include pH, ion strength of water, types of membrane, operation pressure, and the membrane state [15-17]. The operators in treatment plant may not caution the differ of trace elements removal, and some mechanisms are not very clear on the complex background of coal mine water. As the trace elements are very important on the surrounding water bodies, the water treatment may need more carefully evaluation on discharging and reuse.

**Figure 3.** Boxplot of trace element concentrations in water samples (a).

**Figure 4.** Boxplot of trace element concentrations in water samples (b).
3.2. Trace element concentrations in surrounding waters

The carbonate water is likely to be source of coal mine water. And the surface water may be impacted or contaminated by coal mine water. To evaluate the impact of coal mine water on surface water, water samples from both the coal mine district and non-coal mine district were collected.

By dissolution of limestone in the Ordovician system and Taiyuan formation, the carbonate water has equivalent concentrations of Na, K, Cl, and HCO$_3^-$, but much higher Ca, Mg, and SO$_4^{2-}$ ions than coal mine water. Most of the trace elements have much lower concentrations in the carbonate water.

For the surface water, most of the trace elements have lower concentrations than coal mine water. When compared with “Environmental quality standards for surface water”, As, Hg, Se, Pb recognized as type I, however, Cd in the surface water in the coal mine district belonged to type V. Some other metal elements, such as Zn and Fe, showed higher concentrations than that in coal mine water. Although the metal element concentrations were low in the coal mine water, the source of metal elements from the coal mine water could not be excluded. As the coal mine water in this district has a relative high pH, the metal elements are adsorbed by clay, organic matter and other sorbents, or in a mineral form. While environment changes in the surface water, the trace elements may be released. Two types of documents could support this hypothesis: first, former researchers have found the surface water and ground water contaminated by trace elements, and the relationship of surrounding water and coal mine water [20-22]; second, the trace element concentrations in coal mine district and non-coal mine district are significantly different. [Na+K], [Ca], [Mg], [Cl], [SO$_4^{2-}$], [HCO$_3^-$], hardness, and TDS in the coal mine district are 6.3, 1.6, 2.8, 3.6, 2.3, 2.7, 2.1, and 2.9-fold than those in the non-coal mine district. For the important trace elements, [As], [Hg], [Se], [Zn], [Cd], [Sb], [Pb], and [Co] are higher in the coal-mine district surface water, which are 2.3, 1.5, 2.0, 5.7, 21.8, 4.7, 14.1, 3.6 times than surface water samples in the non-coal mine district.

4. Conclusions

Twenty-one water samples were collected in the study area. Concentrations of major ion and trace element of coal mine water, carbonate water, surface water, and treated water were tested. It was turned out that conventional treatment technologies play weak role in the removal of major ions and trace elements. The reverse osmosis technology could remove major ions effectively from the coal mine water.

The reverse osmosis technology performed differ in trace element removal. Most of metalloid elements were partly removed. Some metal elements were rejected effectively, while some others elevated. By comparing with drinking water standard, concentrations of most important trace elements were higher than the highest acceptable value. The factors that may impact the removal efficiency includes operation pressure, pH of coal mine water, membrane type, and membrane state, etc.

The surface water in the coal mine district is probably impact by coal mine water, for the trace element concentrations were much higher than that in surface water of non-coal mine district. Metal and metalloid elements show different behavior, suggesting different reaction path while the coal mine water mixing with surface water.

Because of the potential toxicity of trace elements, the treatment of coal mine water and discharge need more carefully evaluation and technological improvement.

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