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TECHNICAL ARTICLE

Effects of Coalbed Methane Co-produced Water for Irrigation in China's Qinshui Basin: An Experimental Field Study

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Abstract
The management of coalbed methane (CBM) co-produced waters (hereafter refer to as produced water) has broad effects on the native plants and soils in the Qinshui Basin of China and has become a high priority over the past decade. To investigate the short-term effects of produced water on crop production and soil health, a field study was conducted in the Shizhuang Block, which is one of the most active CBM-producing areas in the Qinshui Basin. Spring maize was grown during 2015 under the following flood irrigation treatments: freshwater (as a control), and three produced water sub-treatments with electrical conductivity (EC) values of 1.9 ± 0.9, 2.3 ± 0.5, and 3.4 ± 1.1 ds/m. The SAR concentration of the produced water ranged from 17.62 to 44.45 meq/L; the excessive SAR content makes the water unsuitable for direct irrigation without amendment. The pH, SAR, and constituent concentration of soil samples and crop yield were tested before planting and after harvest, while the soil moisture and plant height were measured monthly from June to October. The growth parameters indicated that salinity decreased total crop yield and individual plant height. The monitored soil indexes showed that the SAR content at a depth of 20 cm (top soil) exceeded that at 40 cm, and increased as the EC of the irrigation source increased at both depths. However, there were no obvious differences in pH or chemical composition of the soil samples, all being less than the Republic of China's soil pollution standards. Soil moisture was determined more by the monsoon climate than by changes in the soils' hydraulic properties.

Keywords Soil property · Crop performance · Salinity · SAR

Introduction

Coalbed methane (CBM) is formed in confined coal formations through complex geological process, where it remains trapped by water pressure. There are ≈ 240 trillion m^3 of CBM around the world buried at depths less than 2000 m, making it an important source of energy for residential and industrial sectors (Bao et al. 2014; Li 2006). China, with the third largest national CBM deposits in the world with 36.8 trillion m^3 (Chen and Hu 2000) is poised to use CBM as a clean, plentiful fuel that can supplant coal and oil (Bao et al. 2013; Md and Daigoro 2008).

CBM is extracted from coal seams by pumping water from wells installed in coalbeds (Bryner 2004; Ingelson et al. 2006). This pumping decreases the hydrostatic pressure, allowing the methane gas to flow up to the wellbore surface, where it is more easily extracted (U.S. Geological Survey 2000). The quality and quantity of produced water vary widely by production phase and coalbed geologic features, scaling from different basins to individual wells within the same basin (Kędzior 2009). According to different production phases and regional water quality characteristics, the produced water in China can be classified into four types: improved water, produced water containing trace elements, fluoridated produced water, and highly saline produced water (Sang et al. 2009). At present, most produced water China is high in salinity; for example, the salinity of CBM-produced water in the Zaoyuan production area of

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the southern Qinshui Basin is approximately 58,000 mg/L (Wang et al. 2009).

Obviously, managing the enormous quantity of produced water is a significant challenge for it has a wide range of potential environmental and social impacts, including surface disturbances, water resource depletion and contamination, and impacts to wildlife, aquatic organisms, and aesthetics (Stearns and Tindall 2005). The USA, the first country producing CBM commercially, has a rich experience in disposal of produced water. These produced water management methods include aquifer recharge, surface discharge, land application disposal, impoundments, agricultural use, industrial use, and domestic and municipal use (Boysen et al. 2002; Ganjegunte et al. 2005). For countries like China, where CBM production developed later, produced water disposal methods are immature: produced water is usually discharged into lined or unlined impoundments, which is a waste of the water resource and a potential source of contamination.

Several areas in China with abundant CBM deposits are located in arid or semiarid regions (Valipour 2017), where water shortage is a major factor limiting sustainable agriculture. Direct discharge of produced water to surface soils could potentially supplement precipitation during the normal growing season in these regions. However, if the produced water contains high sodium adsorption ratio (SAR) levels (high concentrations of sodium relative to calcium and/or magnesium, along with high amounts of bicarbonate), the water may degrade or deflocculate clay particles within the soils, thereby reducing the water absorption capacity of soil, adversely affecting vegetation (Bergquist 2003; McBeth et al. 2003; Reddy et al. 2001).

Soil health and crop yield are two important indicators of concern when using saline water for irrigation (Wang et al. 2015). Research on these problems have mainly focused on secondary treated sewage or distribution of salinity (Bern et al. 2013; Hu et al. 2013; Johnston et al. 2014; Lado et al. 2012; Valipour 2013) and there has been little research on the effects of using produced water for irrigation in China. Information on how produced water irrigation affects the water-soil environment may help in assessing the feasibility of using produced water for irrigation. In this study, we conducted a short-term field experiment on the use of produced water for watering spring maize to identify the potential impact of produced water on soil and plants.

Materials and Methods

Study Area

The Qinshui Basin, located in southeastern Shanxi Province, is surrounded by the Taihang, Taiyue, and Zhongtiao Mountains at geographic coordinates: 35°24′–36°04′ north latitude, longitude 115°55′-112°47′. Several companies, such as China United Coalbed Methane Corporation (CUCMC), China National Petroleum Corporation (CNPC), and Lanyan Coalbed Methane Co., are extracting CBM in the Qinshui Basin, making it one of the most active basins in China for CBM exploration. The latest evaluation shows up to 3.98 megatons of CBM at depths shallower than 2000 m (Su et al. 2005).

The study area is located in the Shizhuang region of the southern Qinshui Basin, in an area owned by CUCMC (Fig. 1). The Shizhuang region is located in a temperate continental monsoon climate zone; it is hot and rainy in the summer, with temperatures averaging 20.7–24.8 °C, while the winter is cold and dry, with average temperatures of − 3.1 to 6.5 °C. Summer droughts are frequent. The soil in the area is cinnamon in colour, and millet and maize are the main grain crops. Further climate details during this study are given in supplemental Table S-1.

The area under study is the southeastern limb of the Qinshui Basin's complex syncline, with the coal-bearing strata dipping about 5°NW, and the structures within the studied area are simple. The main coal-bearing strata include the Permian Shanxi and Carboniferous Taiyuan formations (Su et al. 2005). The hydraulic aquifers connected to the coal seam in the study area include a sandstone aquifer in the Shanxi Formation and a limestone aquifer in the Taiyuan Formation, neither of which outcrop within the studied area. These two formations are separated from the two aquifers by an aquiclude layer, with a weak hydraulic connection. In general, groundwater recharge of the aquifers within the studied area show is dominated by slow inter-layer runoff; the area of discharge is not obvious. Both formations outcrop in the eastern Qinshui Basin beyond the studied area, where they are recharged by precipitation, as shown in supplemental Fig. S-1.

Methods

Studies were carried out in 2015 (March–October) in the Shizhuang region. Five plots were constructed, each 4 m^2 (2 × 2 m), confined by plastic film placed in the soil at a depth of 40 cm. All four plots were planted with maize, and the plant density was five plants per m^2. The irrigation waters were divided into two categories: CBM water from three different CBM production wells (plots A, B, and C) and fresh water from the surrounding river (plot D). Each plot was irrigated once a week with 360 L of the above-mentioned types of water in addition to natural precipitation. Plants in plot E were grown using natural precipitation only (detail could be seen in supplemental Table S-1).

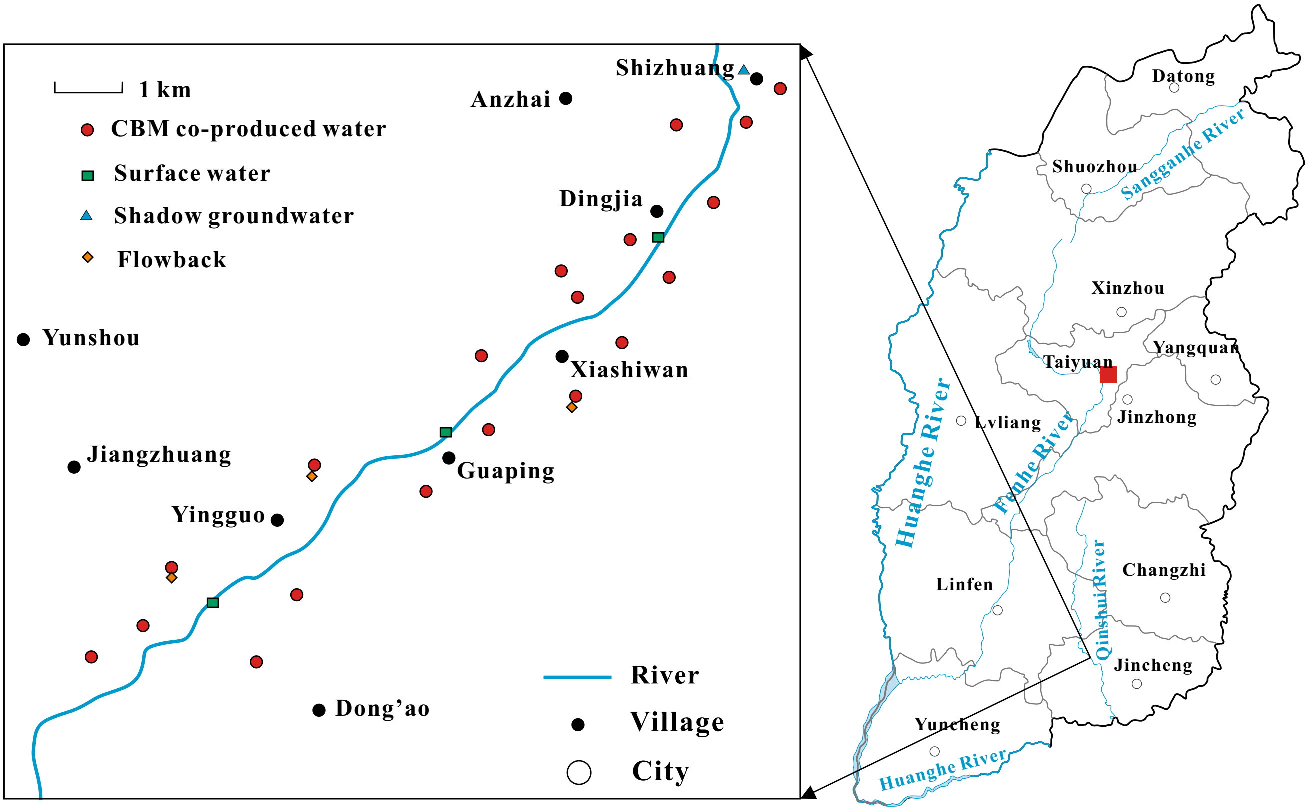

Fig. 1 Shanxi province and sampling sites

The CBM water used for irrigation was discharged into a holding pond and analyzed for pH, major ions (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-}, HCO_3^-, F^-), and trace constituents (Zn, As, Cu, Pb, Mn, Fe, Cd). In each plot, a soil auger was used to randomly collect four replicate soil samples at a depth of 0–20 cm and 20–40 cm monthly and after the irrigation season, and then the averages were calculated. To ensure that the soil samples were representative, we adopted a grid method for sampling. Samples were transported to the laboratory to test the parameter values, including pH, analyzed constituents, SAR, and moisture content.

The pH and electrical conductivity (EC) values for CBM water and soil samples were determined using pH and EC electrodes (Rhoades 1999; Thomas 1999). Total dissolved solid was measured using the gravimetric method, by calculating the difference between before and after the water samples were dried at 105 °C. Soil samples were stored in polyethylene bags to prevent moisture loss during transportation to the laboratory. Subsamples were dried at 105 °C until a constant weight was obtained (about 10 h), and soil moisture contents were determined using the difference between the field-moist and oven-dry weights. Metal and semi-metal content was determined using atomic absorption spectroscopy (AAS) after digestion in concentrated nitric acid. The solutions were filtered through Whatman's No. 42 paper, up to a standard volume, and analyzed for metals. Water samples were acidified using nitric acid and analyzed using the flameless AAS method. Major ions in soil and water samples were analyzed with inductively coupled plasma spectrometry. The SAR of the CBM water and soil samples was calculated (Li et al. 2013a, b):

$$SAR = [Na^+]/[Ca^{2+} + Mg^{2+}]^{1/2},$$

where [ion] represent milliequivalent concentrations (meq/L) of the respective ions.

In the different growth phases of the maize crops (seedling stage, booting stage, and kernel stage), metal content was analyzed using the AAS method. Before analysis, the plant samples were digested by thoroughly washing them three times in deionized water, drying them at 60 °C, and a wet-ash treatment in concentrated nitric acid. From June to October, plant height was measured monthly to evaluate the impact caused by the CBM water. At harvesting season, maize crops were harvested from each plot and weighed to estimate yield.

Results and Discussion

CBM-Produced Water Characteristics

Water samples were taken in February 2015, and a Piper plot was drawn (supplemental Fig. S-2). The pH values of the water samples from different sources in the study area were not distinguishable, ranging from 7.6 to 8.5 (weakly alkaline). The total dissolved solids (TDS) of the water co-produced with the CBM ranged from 678.1 to 1621.7 mg/L, with an average of 1144.2 mg/L, and Na^+ was the main cation, accounting for 85.2–97.7% of the total cations, with an average of 93.1%. HCO_3^- was the main anion, accounting for 47.4–96.4% of the total anions, with an average of 80.3%. In some water samples, the Cl^- concentration was also high, accounting for 39.6% of the total anions. The water type was mainly HCO_3-Na type, or Cl-HCO_3-Na type, which is consistent with Van Voast (2003) research results and quite different from the water produced in conventional oil and gas fields.

The TDS of the surface water is similar to that of the shallow groundwater in the study area, with a TDS of 307.2–353.5 mg/L, and an average of 331.5 mg/L. Ca^{2+} is the main cation, accounting for 59.7–72.4% of the cation total. HCO_3^- is the main anion, accounting for 74.8–78.3% of the total anions. The water type is mainly HCO_3–Ca.

In deciding whether the CBM-produced water is suitable for farmland irrigation, the most important indicator is the risk of salinization and alkalization (Smith et al. 2009). In the USA, water with an SAR value in the range of 6–12 meq/L is considered suitable for irrigation (Brinck et al. 2008). The SAR of the produced water in the Power River Basin of the United States ranges up to 70 meq/L (McBeth et al. 2003), which greatly affects the use of produced water. The EC and SAR of the produced water in the southern Qinshui Basin are plotted in Fig. 2.

According to the US Salinity Laboratory, the SAR and salinity should be considered simultaneously to determine if a particular water quality is suitable for irrigation (Li et al. 2014, 2016a, b; Richards 1954; Wu and Sun 2016). Salinity is expressed in terms of EC and divided into four categories (Richards 1954): low in salt (C1), salty (C2), highly saline (C3), and extremely saline (C4). The SAR value of the water is also divided into four categories: low (S1), moderate (S2), high (S3), and extremely high sodium content (S4).

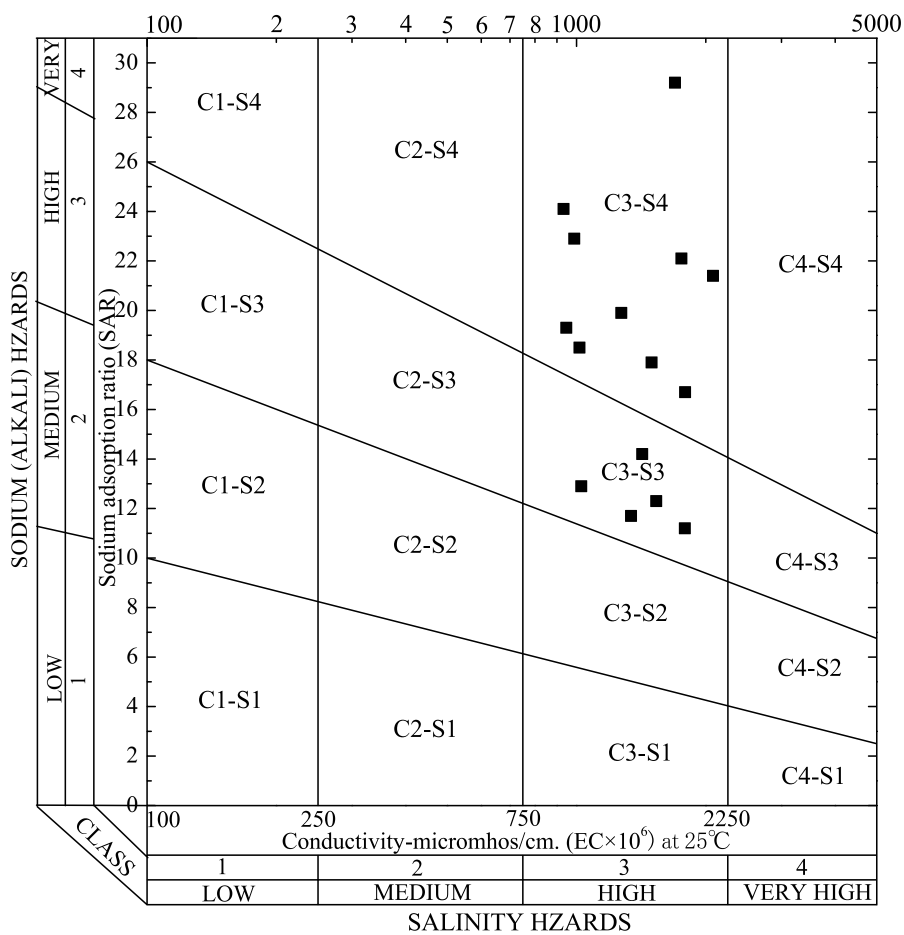

Fig. 2 Diagram for the classification of irrigation waters

Table 1 Some chemical indices of water used for irrigation

Parameter	pH	EC (ds m^{-1})	TDS (mg L^{-1})
Freshwater	8.0 ± 0.1	1.2 ± 0.4	300 ± 100
Well A	8.8 ± 0.4	2.3 ± 0.5	600 ± 200
Well B	8.5 ± 0.6	1.9 ± 0.9	1000 ± 200
Well C	8.7 ± 0.3	3.4 ± 1.1	1700 ± 500

The produced water in the study area is mostly C3–S3 and C3–S4, namely, highly saline and high in sodium and highly saline with extreme sodium levels. The water cannot be used to irrigate the soil directly and must be treated; otherwise, it will cause significant damage to the soil.

Effects of CBM-Produced Water for Irrigation

Effects of CBM-Produced Water Irrigation on Soil Properties

Some chemical indexes of coproduced water from three different CBM production wells used for irrigation are shown in Table 1, revealing low, medium, and high TDS concentrations. Concentrations of Na^+, HCO_3^-, and Cl^- in water samples from well C were significantly higher than in water from the other two wells. This may be due to the production period, as other data in this study indicate that concentrations of Na^+, HCO_3^-, and Cl^- decrease with mining time. Well B has been in production for a long time, so the variations of ionic concentrations in water from well B have become less pronounced and more balanced. Unlike the produced water, the water from the nearby river is dominated by Ca and Mg. Moreover, the fresh water sample contains more SO_4^{2-} than water samples from wells A and C, possibly due to manmade environmental pollution. Concentrations of other ions in the fresh water are significantly less than in the CBM water.

Constituent concentrations were relatively low in both the fresh and produced water, except for the As, Pb, and Mn in water samples from well C (Table 2). Possibly due to the alkaline environment, a large portion of soluble metals in the CBM water could have precipitated from the liquid phase into the solid phase (slime) (El-Hasan 2006). The special case of well C may be the result of human activity, for example, contaminated by well drilling or hydraulic fracturing.

The short-term use of produced water for irrigation did not significantly increase metal concentrations in the soil (Table 3), since these elements were relatively low in all of the water types used in this investigation. The metal concentrations of the soil samples from plot D, irrigated by fresh water (as a control), were not statistically different

Table 2 Content of water used for irrigation (mg L^{-1})

Parameter	Zn	As	Cd	Cu	Fe	Mn	Pb
Freshwater	ND	ND	ND	ND	ND	ND	ND
Well A	1.11 ± 0.18	0.0039 ± 0.003	0.014 ± 0.005	0.4 ± 0.09	0.051 ± 0.016	30.4 ± 5.2	< 0.1
Well B	1.06 ± 0.27	0.0026 ± 0.001	0.012 ± 0.007	0.98 ± 0.02	< 0.05	4.67 ± 1.1	< 0.1
Well C	ND	7.50 ±	0.73 ± 0.19	< 0.06	0.18 ± 0.07	7.20 ± 1.5	12.30 ± 2.9

Table 3 Constituent concentrations in soils irrigated with different types of water

Parameter	Sampling depth (cm)	A	B	C	D	E
Zn (mg kg^{-1})	0–20	52.33	44.59	52.76	59.43	69.98
	20–40	52.33	42.12	59.18	61.03	66.87
As (mg kg^{-1})	0–20	5.67	6.00	11.06	5.40	9.25
	20–40	5.98	5.98	10.02	5.40	9.10
Cu (mg kg^{-1})	0–20	17.98	20.06	18.82	17.43	20.00
	20–40	17.34	18.03	19.12	16.01	21.03
Pb (mg kg^{-1})	0–20	16.88	15.02	23.14	16.88	15.80
	20–40	17.00	14.31	23.05	17.00	14.78
Mn (mg kg^{-1})	0–20	584.42	561.33	542.11	534.12	554.00
	20–40	580.12	556.00	543.00	529.11	554.00
Fe (%)	0–20	4.043	3.697	4.142	3.969	4.000
	20–40	4.100	3.68	4.100	4.085	3.721
Cd (mg kg^{-1})	0–20	ND	ND	ND	ND	ND
	20–40	ND	ND	ND	ND	ND

from that of soil samples from the three plots (A, B, and C) irrigated with produced water. Although water from well C had increased the Pb and As content of the soils, their levels were still safe based on the environmental quality evaluation standards for farmland of edible agricultural products (HJ/T332-2006). All constituents were under or only slightly more than that of the Shanxi Province background soil and scored as Grade I according to the environmental quality standard for soils (GB15618-1995).

The soil samples collected at the end of the irrigation season showed higher pH and SAR values in all of the produced water irrigated soils (plots A, B, C) compared to the fresh water irrigated soils (plot D), according to Figs. 3 and 4. The increase in pH varied with the type of water used for irrigation: the pH of the soils in plot C, which was irrigated by the high TDS produced water, had noticeably increased. Soil pH in the 0–20 cm soil layer was significantly greater than in the 20–40 cm soil layer, except in plot E, whose pH value was 8.3 in both the top soil and subsoil. Obviously, the produced water irrigated soils (plots A–C) had greater SAR values than the fresh water irrigated soil (plot D). The increased SAR values in soils was due to import and mobilization of Na^+ from the produced water. The SAR values of soil in plot C were significantly greater than those of other plots at depths of 0–20 and 20–40 cm, as the water source for this plot had the highest concentration of Na^+, EC, and TDS.

The soil irrigated with the produced water contained some neutral or slightly alkaline salt, such as NaCl, $CaCl_2$, Na_2SO_4, and $MgSO_4$, which accumulated at the soil surface, increasing the alkalinity. Soil is considered salty when this accumulation is elevated enough to affect plant growth. When Na^+ gets into soil, the soil becomes alkaline due to the presence of HCO_3^- and CO_3^{2-}. Alkalinity usually accompanies salinization. The more serious the sodification, the less the soil aggregate (Tang 2004). Soils with EC

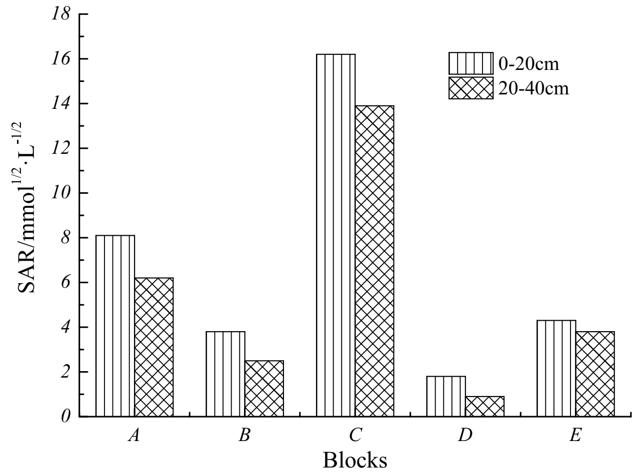

Fig. 4 Mean soil solution SAR values

> 4 dS m^{-1} and SAR < 13 $mmol^{1/2}$ $L^{-1/2}$ are considered saline, whereas soils with EC > 4 dS m^{-1} and SAR > 1313 $mmol^{1/2}$ $L^{-1/2}$ are considered saline-sodic soils (Chhabra 2005; Gupta and Abrol 1990). Therefore, the soil in plot C that had been irrigated with higher salt CBM water was seriously saline-sodic. We also found that in plot E, which was not irrigated with fresh or produced water, SAR and pH values were higher than in plot B and plot D. This may be due to the water quality and accumulated salinity due to the local drought climate (Figs. 3, 4).

During the experiment, the soil moisture ranged from 32 to 45% in the 20–40 cm layer, and from 22 to 33% in the 0–20 cm layer. Obviously, the soil moisture content in the subsoil was higher than in the top soil in all of the plots (Fig. 5); evaporation of water from the top soil layers presumably caused the uniform changes. Overall, the soil moisture contents first increased and then decreased, basically

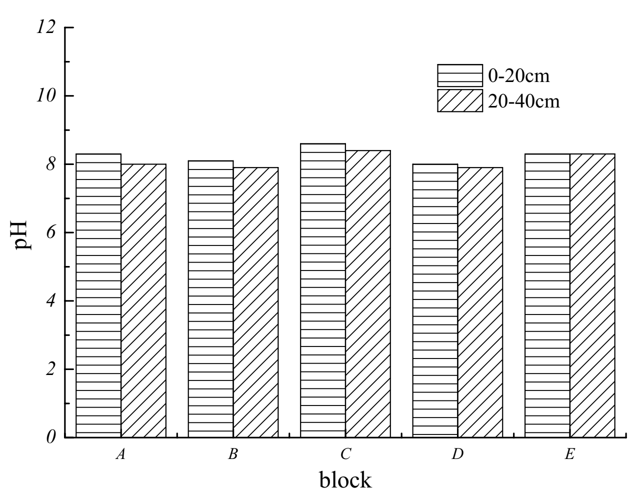

Fig. 3 Mean soil solution pH values

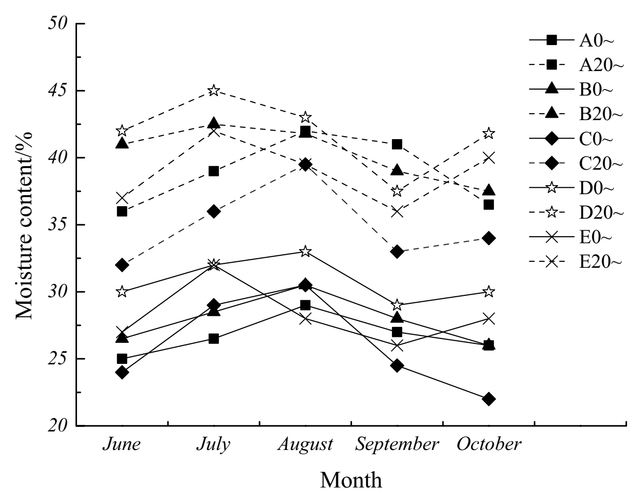

Fig. 5 Mean soil moisture content during different months

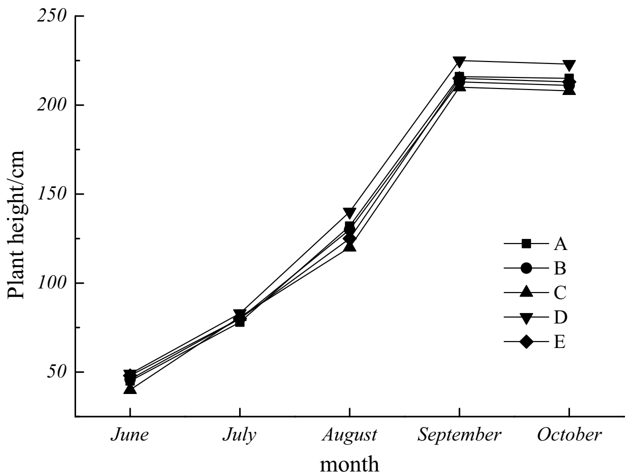

Fig. 6 Status of maize growth

peaking in July or August. This result was determined by the local monsoon climate (supplemental Table 1). In addition, the individual water source for irrigation affected the native hydrotropism, since higher SAR values will decentralize soil particles, decreasing water retention (Bardhan et al. 2016; Valipour and Montazar 2012), and cause serious soil erosion. Plot E, which was not irrigated by produced water or fresh water, also had a low soil moisture content. Drought caused the soil moisture content to decrease and the salinity to increase. In brief, the combination of rain and heat, plant growth pattern, and the salinity of irrigation water, all affected the distribution of soil moisture content.

Effects of CBM Produced Water Irrigation on Spring Maize Growth

Figure 6 shows the plant height for the different irrigation conditions. It can be seen that plant height varied in an "S" curve, though the changing trends in spring maize plant height under different irrigation conditions were essentially identical; however, the specific plant height were significantly different for the different irrigation waters. From June to October, the spring maize plant heights in plots A-E ranged from 40 to 50 cm in June, 78–87 cm in July, 123–140 cm in August, and 210–223 cm in September and October. According to Fig. 6, the best growth was in plot D, which was irrigated by fresh water. Although the plants grown in plots A and B did not grow as well as in plot D, the gaps were small, sometimes irrelevant, such as plot B in June. The spring maize in plot C grew slowly. Thus, salinity is one of the main environmental factors affecting plant growth and development. Crops in plot E with non-irrigative activity were worse than ether plot D (irrigated by fresh water) or plots A and B (irrigated by relatively low saline produced water). Irrigation water from well B contains more SO_4^{2-} than the other irrigation water, but there was no visible impact on maize growth; biochemical changes in the soil may have led to this result.

Supplemental Fig. S-3 shows the spring maize crop yield results. It is evident that produced water had an inverse relationship with crop yield, as an increase in TDS from an average value of 800 mg L^{-1} for the fresh water to 600, 1000, and 1700 mg/L from wells A, B, and C, respectively, for the produced water (Table 2) reduced the crop yield by 17.4, 10.9, and 30.4%, respectively, in the corresponding plots. But the plants in plots A and B achieved an 8.5 and 17.1% increase, respectively, relative to plot E. This outcome is in line with previous research showing that in comparison with dry land, the crop yield increased from 10 and 30%, topping out at 49% in plots treated with saline water. Maize crop yield in plot C was the lowest, for the water was too salty for the plants.

Plant analyses indicated few metals or semi-metals in the maize crop (seedling stage, booting stage, and kernel stage), due to the low metal concentrations in the soil and in the water used for irrigation (Table 4). This could also reflect the nature of maize plants since the transportation of metals is difficult in tall plants (Minkina et al. 2001; Rimawi et al. 2009). In this study, the detection range was limited to the leaves of the maize; the roots and seeds were not examined, so metals may have been enriched in those areas.

Conclusions

Although the CBM produced water was more saline than the fresh water, produced water has a potential utilization value for irrigation in arid and semiarid areas. Using produced water as a short-term irrigation source did not significantly increase the soil's metal content because the concentrations of these elements were relatively low. Plants and soils were apparently not adversely affected by metals due to irrigation with the produced water. However, it did affect soil pH and SAR, as the soil samples collected at the end of the irrigation season had higher pH and SAR values in all of the produced water irrigated soils, though soil salinity also increased due to drought conditions. Salinity is one of the main factors affecting plant growth, but the maize crop yield had an inverse relationship with CBM co-produced water. Irrigating with high salinity CBM produced water reduced crop production relative to fresh water, but in the dry season, irrigation using produced water with an appropriate salt content can help increase crop yield.

Table 4 Concentrations of Zn, As, Cu, Pb, Mn, Fe, and Cd for maize plants in different plots

	Growth phases	A	B	C	D	E
Zn (mg/L)	Seedling stage	0.02	0.07	0.04	0.01	0.04
	Booting stage	0.05	0.08	0.06	0.05	0.07
	Kernel stage	0.08	0.09	0.10	0.11	0.09
As (mg/L)	Seedling stage	ND	0.01	ND	ND	ND
	Booting stage	ND	0.02	ND	ND	ND
	Kernel stage	ND	0.04	ND	ND	ND
Cu (mg/L)	Seedling stage	ND	ND	ND	ND	ND
	Booting stage	ND	ND	ND	ND	ND
	Kernel stage	ND	ND	ND	ND	ND
Pb (mg/L)	Seedling stage	ND	ND	ND	ND	ND
	Booting stage	ND	ND	ND	ND	ND
	Kernel stage	ND	ND	ND	ND	ND
Mn (mg/L)	Seedling stage	0.03	0.02	ND	ND	ND
	Booting stage	0.07	0.08	ND	0.01	0.02
	Kernel stage	0.09	0.12	ND	0.04	0.07
Fe (mg/L)	Seedling stage	0.11	0.43	ND	0.88	0.21
	Booting stage	0.39	0.80	0.05	1.20	0.65
	Kernel stage	0.98	1.05	0.54	1.50	0.77
Cd (mg/L)	Seedling stage	ND	ND	ND	ND	ND
	Booting stage	ND	ND	ND	ND	ND
	Kernel stage	ND	ND	ND	ND	ND

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