Geochemical and isotopic evidence of shallow groundwater salinization in a reclaimed coastal zone: the Yellow River Delta, China

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Abstract This paper aims to improve the current knowledge of groundwater salinization processes of a young coastal aquifer in the Yellow River Delta (YRD) and to assess the impact of both the transfer of irrigation water from the Yellow River and seawater intrusion, using a combination of hydrochemical–isotopic measurements. The major ion/ Cl^- ratios generally closely follow the mixing line between freshwater end-member (the Yellow River) and saline water end-members (salt groundwater or seawater), indicating the importance of mixing under natural and anthropogenic influences. The examination of stable isotope in groundwater (ranging from -9.0 to -3.5 ‰ for $\delta^{18}\text{O}$ and from -73 to -29 ‰ for δD , respectively) and chloride concentrations (~ 2 to 629 meq/L) confirms that the saline end-member is associated with saltwater rather than present seawater, and most groundwater samples are plotted on mixing lines between the Yellow River and saltwater. The similar range in stable isotopes ($\delta^{18}\text{O}$ and δD) and Cl^- compositions between saltwater and confined aquifer supports the

hypothesis that upward seepage of confined saline water intruded the shallow aquifer. Moreover, the deviations in some cases from mixing line imply water–rock interaction is also important in dominant control on groundwater chemistry, such as cation exchange, SO_4^{2-} reduction, and NaCl solution. The brackish water was characterized by slight excess of Na^+ and deficit of Ca^{2+} compared to non-reactive mixing line. This indicates that the coastal aquifer is generally freshening with respect to extensive irrigation network, as well as the transfer from the Yellow River for ecological recharge into wetland.

Keywords Deltas · Irrigation water · Water mixing · Salinization and freshening · Geochemistry · Stable isotopes

Introduction

Groundwater salinization in coastal areas is becoming a limiting factor for agricultural and further economic development in arid and semiarid regions (Farid et al. 2015; Syvitski et al. 2009; Vengosh et al. 1999). The associated increase in total salinity can eventually render freshwater resource unusable (Arfib and Charlier 2016; Oh and Kim 2016). Potential salinization sources are diverse, such as seawater intrusion and upward flow from deep saline water trapped in the sediments (Han et al. 2011), the dissolution of secondary minerals within the sedimentary formations (Howard and Lloyd 1983), return flow from irrigation water (Carol et al. 2009; Cruz-Fuentes et al. 2014), anthropogenic contamination, or most often a combination of some of these processes (Ghabayen et al. 2006; Ledesma-Ruiz et al. 2015; Marimuthu et al. 2005; Park et al. 2005). Coastal aquifers are thus generally

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complex, due to the transient state influenced by continual modifications of freshwater and saltwater interface: sea-level oscillations, extreme climate, and water table variations due to anthropogenic and/or natural variations in the water balance.

The YRD represents a prime example of coastal area vulnerability, where the equilibrium between human activities and natural evolution has always been fragile. The coastal aquifer was developed over three marine transgressions and regressions since Late Pleistocene. Its flat morphology reveals the joint influence of tidal dynamics on sediment input from inland rivers and coastal transport, which determined the hydrogeological as well as hydrochemical processes. Since 1980, with the agricultural development and exploitation of land in the YRD, irrigation has depended entirely on water transferred from the Yellow River. In 1986, 1991, and 1999, the volume of water transferred totaled 3.2, 4.6, and 5.3 million m³/a, respectively (Gonçalves et al. 2007). Extensive irrigation without adequate drainage has caused rising groundwater levels and disturbed the preexisting water–salt balance in the soil zone and in the shallow aquifer. Alternatively, coastal wetland plays a significant role in the equilibrium of regional ecosystems, and modifications in groundwater flow regimes that can lead to increased salinization of wetlands when groundwater contains more salt than surface water (Négrel et al. 2003). It is thus necessary to discuss how to control the dynamics of rising groundwater levels and salinity, and to identify the origin of salts for both water management and remediation in this salinized and water-deficient region.

Many eco-environmental problems observed in the YRD, such as degradation of vegetation ecosystems, desertion of farmlands, and soil salinization, are closely related to groundwater salinization. Although the saline groundwater is being replaced by freshwater in the lower reaches of the river and in the delta area, through irrigation and seepage from the Yellow River, the salinity of groundwater is still an imperative issue in the delta, due to the interface between freshwater and saline water which has always been precarious under the natural and anthropogenic influences.

A number of studies have used either GIS or remote sensing models of the Yellow River Delta to investigate the influence of groundwater salinity on soil salinization (Fan 2007; Guan 2001a; Lou et al. 2007), and pollution of hydrocarbons, nitrates, polychlorinated biphenyls (PCBs), trace metals, and polycyclic aromatic hydrocarbons (Li et al. 2015; Song et al. 2013; Xing et al. 2005). In previous hydrochemical investigations, distribution of salinity patterns in the aquifer was investigated together with geochemical process (such as base-exchange reactions, mineral dissolution/precipitation) based on major ion

chemistry (An et al. 2012; Yuan 2006). The present research study combines field surveys of hydrochemistry (major and minor ions) and stable isotopes (δD and $\delta^{18}\text{O}$) to understand the cause and mechanisms of salinization in relation to recharge, origin of salinity, mixing process, directional processes (salinization or freshening), and water–rock interactions. The results will contribute to sustainable water and land management, as well as to preventing quantitative and qualitative alteration of environmental conditions.

Study area description

The YRD was selected between 37° 26′–38° 10′N and 118° 21′–119° 20′E (see as in Fig. 1). This area was characterized by a monsoon climate of the warm temperate zone. The average annual precipitation was 530–630 mm, of which 70 % was occurring during summer (June–September), while the average evapotranspiration is 1900–2400 mm, indicating an evaporation/precipitation ratio close to 3.5 (Li et al. 2014; Yu and Liu 2012). The highest elevation of the delta was approximately 13.3 m above mean sea level in the west; the lowest elevation was less than 2 m along the coast. The average topographical gradient was approximately 1/8000–1/2000 (Cheng et al. 1987; Xie et al. 2015; Zhao et al. 2015). The elevation of the YRD reflected the sedimentation history of the shifting river channels. The high sediment load transported from the whole basin contributed to the enlargement of the delta and upward rising of the riverbed, which caused the YRD region expanding in area approximately 20–25 km²/year (Wu et al. 2015; Ye et al. 1997).

The YRD was located in the southeast of the young depression zone of the North China Plain, mainly under the control of New Cathaysian structure belt and northwest direction structure belt (Wu et al. 2004; Zhou et al. 2016). The cross sections A–A′ and B–B′ (see Fig. 1) run from the southeastern coast (Huanghai Bay) to the northern coast (Bohai Bay) and from the west to the east, respectively, reflecting the characteristic deposits and lithology of the aquifers. The aquifers mainly consisted of riverbed and estuary sandbar facies, and the riverbed overlaid the estuary sandbar, formed by riverbed deposits during the different periods. The base of cross section A–A′ was a sandy clay layer. The top of this layer varied from 7 m below sea level in the south to 20 m below sea level in the north. Over this base laid a thick layer of sandy clay with a typical biconvex shape. These bodies also corresponded with the axes of the abandoned river course, and the elder sand bodies were overlain by younger backswamp deposits (mostly sandy clay), or by river levee deposits closer to the path of the younger river. The B–B′ cross section showed

Fig. 1 Location of sampling sites in the Yellow River Delta (a), and geological cross-sectional maps along A–A' and B–B' (b)

the sediment layers in the recent delta of the Qing Shui Gou course: the present mouth of the Yellow River. The main aquifers in the YRD were porous Quaternary sediments, which filled the delta with a thickness ranging from 8 to 20 m, and the depths of aquifers increased from the mainland to shore (Xue et al. 2000). Brines have been detected up to 10–20 km from the coast and distributed in a depth of 30–50 m (Fan et al. 2012; Yao and Yang 2007). The minerals in the aquifer included 62 % illite, 17 % koalinite and halite, 8 % montmorillonite, and smaller amounts of halloysite, vermiculite, etc. (Weng et al. 2008;

Yu 2002). The non-clay minerals were quartz, plagioclase, orthoclase, mica, amphibole, calcite, etc. (Saito et al. 2001; Han et al. 1996).

The aquifers are characterized by a complex multilayered framework (Fig. 1). Close to the inland, shallow and deep groundwater is unconfined, while toward the coastline, deep groundwater (~at and below 35 m) becomes confined due to increasing occurrence of upper sandy clay. The unconfined aquifer in the delta is recharged via infiltration of precipitation, river and pond leakage, and agricultural return flow (Guan 2001b), and it is mainly

discharged by evapotranspiration in the shallow soil zone and fluxes of groundwater to the surface river, to the ditches, or to the Bohai Sea (Liu et al. 2014; Taniguchi et al. 2008). The soil texture in the unsaturated zone is mainly composed of clay silt and sandy silt, with a loose structure resulting in more permeability, which makes the infiltration (i.e., recharge) and evapotranspiration (i.e., discharge) prone to occur in the vertical direction. Groundwater dynamics in the deep confined aquifer is influenced by climatic and hydrogeological conditions, and particularly the volumes of groundwater exploitation for salt production (Han et al. 2014; Kreuzer et al. 2009). The original salty groundwater has not been suitable for drinking, irrigation, or industrial purposes; therefore, transfer of water from the Yellow River has and will continue to meet domestic, industrial, and agricultural water needs.

Methodology

The field survey was performed at the end of September 2010, covering the entire Yellow River Delta. The sampling campaign was carried out during the period of high flow (the water table was highest based on Fig. 2) in order to denote the largest extent to freshening process over the course of a year. Twenty-three shallow groundwater well samples (at depths of less than 30 m) and twenty-five surface water samples, including two Yellow River samples and one seawater sample, were collected in the old course, abandoned channels, irrigation ditches, surrounding cities and towns, and oil production sites (as shown in Fig. 1).

Temperature, electric conductivity (EC), and the pH value of water samples were measured in situ using a portable WM-22EP, made in Japan. Water samples for cation analysis were immediately filtered through acid-treated Millipore filters (0.45- μm mesh) into polyethylene

bottles, acidified to $\text{pH} < 2$ with HNO_3 (Guaranteed reagent GR, 6 mol/L), and stored at 4 °C until analysis on a Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES, PerkinElmer Co., Ltd., USA). Calibration curves were made after measurements of samples, and data were evaluated by the determination of quality control standards. Reagents, procedural blanks, and samples were measured 6 times in parallel, and the mean of the last three values is reported here. The anions (Cl^- and SO_4^{2-}) were analyzed on filtered samples using a Dionex ICS 900, USA. For both the cation and anion ions, standard reference materials were analyzed as part of the quality assurance and quality control (QA/QC) procedures. The analytical concentrations for standard reference materials were within the range of the certified value, and the replicate analysis was within 10 % variability. Bicarbonate was measured for the filtered samples by titration with H_2SO_4 (0.01 mol/L). Stable hydrogen and oxygen isotopic compositions of water samples were analyzed by the Los Gatos Research Water Vapor Isotope Analyzer DLT-100, USA, with an analytical precision of 1 ‰ for δD and 0.3 ‰ for $\delta^{18}\text{O}$. The δD and $\delta^{18}\text{O}$ values are reported in per mil relative to V-SMOW.

In this study, Piper plot and stiffmap plot are employed to analyze the distribution of facies of water using RockWare Aq.QA version 1.1 for Windows. The saturation indices (SI) are used to identify the hydrochemical processes using PHREEQC version 2.8 for Windows (Parkhurst and Appelo 1999; Han et al. 2014).

Results

Groundwater circulation and water table fluctuation

Surface water transferred from the Yellow River for irrigation in farmland commenced with the beginning of land exploitation and for ecological recharge in wetland dated from 2008, respectively. Due to the gentle topography and extensive irrigation without any drainage systems, the inland groundwater tables have subsequently risen substantially. Figure 2 shows the change in water tables over the course of a year (2010) in some representative wells from different land types (each well depth ranged from 4 to 6 m). Both the water tables in farmland and wetland have sharply risen since July, up to the peak from July to September (the mean water table ranges from ~1.8 to 0.5 m in the farmland and the mean even up to subsurface 0.2 m in the wetland), and gradually fall to the lowest depth from October to the next February. On the contrary, the water table in coast generally shows minimal variation, ranging from 0.75 to 1.2 m in G12 and 2.3 to 2.75 m in G1, respectively. The distinct differences in water table

Fig. 2 Variations in depth to groundwater table (2010) from typical wells in farmland, wetland, and coast regions

between sample sites G1 and G12 can be attributed to the different kinds of land use (Fig. 1), which implies the influence of oilfield water injection occurring in the aquifer of oilfield near the coast. The variation in water table depths over this period is greater in the farmland and wetland regions than that in the coast, indicating an attenuation influence of irrigation water, with water table increasing during and immediately after this period. The response in the coastal wells is however minimal variable, which could be explained by (1) the small degree of influence from irrigation return flow and/or (2) the sea level probably acting as a roughly constant head boundary.

Hydrochemistry

Salinity distribution and water type

The mean total dissolved solid (TDS) content of the Yellow River sampling sites (YR1 and YR2) is 0.66 g/L as calculated from the sum of all major ions analyzed (Table 1), with Ca-HCO₃ type representing the freshest water resource. The other types of surface water, like samples collected from the tributaries, abandoned river channel, irrigation ditch, and pond, are mainly characterized by Na-HCO₃, Na-SO₄, and Na-Cl type, and more salty than the Yellow River, with a TDS value of ranging from 0.8 to 20.0 g/L (mean = 8.4 g/L). Generally, the concentrations of major ions in the groundwater are notably higher than those in the surface water, especially compared to the Yellow River. The TDS of groundwater ranges from 0.51 g/L in the south to 63.91 g/L in the north, with a mean and standard deviation of 8.44 ± 12.47 g/L. Freshwater is mainly found along the modern Yellow River course (G7, G8, G10, G15, and G22). The brackish water characterizes almost the entire coastal aquifer with an average value of 28 g/L and while the saltwater presents a TDS value higher local seawater (~30 g/L) distributed along the coast strip.

As clearly shown in the Piper diagram (Fig. 3), most groundwater samples are concentrated within the central part of the Piper diagram, reflecting the absence of any hydrochemical dominance and indicating the mixing of Ca-HCO₃ and Na-Cl waters. The chemical results enable us to discriminate different water types. Ca-HCO₃-type (G22) freshwater was only found in regions close to the Yellow River (Fig. 1) where the rapid infiltration of fresh river has been noted and other freshwaters change from Ca-HCO₃ type to Na-HCO₃ type, whereas brackish water ranging from Ca-Na-Cl-HCO₃ type to Na-Ca-Cl type, and then to Na-Cl type is located mostly in the transient zone between inland and coast stripe. The Na-Cl facies saltwater samples (G1, G2, G3, G12, G13, and G14) were sampled

from wells located close to the coast where the water table lay above sea level.

Hydrochemical evidence of mixing

In order to discuss processes driving the chemical composition of groundwater, a set of ions ratios are considered to identify mixing mechanisms of fresh and saline groundwater from additional chemical reactions (Giménez and Morell 1997; Yang et al. 2012). The relationship between Cl/HCO₃ molar ratios and Cl⁻ shows a predominantly linear pattern, suggesting a mixing process between freshwater (the Yellow River) and seawater (Fig. 4a). This indicates that mixture of seawater or saltwater is one of the processes responsible for salinization of these groundwater because the samples with lower ratios can be defined as freshwater (Kim et al. 2003), although the deviation of some samples from this line showing that other processes can also be involved. The relationship between SO₄²⁻ and Cl⁻ concentration depicted in Fig. 4b suggests that most data points of freshwater and brackish water show greater dispersion and lower concentration than expected (points lying below the mixing line), while the saltwater samples are plotted near the seawater composition, slightly under the mixing trend.

The relationship between the Na⁺ and Cl⁻ is characterized by a behavior which more closely follows the blending process than that of Ca²⁺ and Cl⁻ (Fig. 4c). The freshwater and brackish water are characterized by relative high Na/Cl ratios up to 2.3 while the saltwater has Na/Cl ratios that are below the value of seawater (Fig. 4d). The relationship between Ca²⁺ and Cl⁻ concentration indicates a substantial correlation among freshwater, brackish water, and saltwater samples, where the freshwater and brackish water are distributed below the mixing line, whereas the saltwater plots above the mixing line (Fig. 4e). The Ca²⁺ depletion in Ca-Cl correlation plot indicates that Ca²⁺ was probably moved by precipitation of calcite and cation exchange in the freshwater and brackish water while the concurrence of the Na⁺ and Cl⁻ values may be attributed to the dissolution of halite which is abundant in this alluvial aquifer. Among the major ions, magnesium can be identified as representative of seawater occurrence at the sub-surface, since natural groundwater seldom consists of similar concentrations (Hem 1985). The relationship between Mg²⁺ and Cl⁻ indicates the Mg²⁺ content of the saline water was consistent with the mixing line, although some deviations occurring in freshwater and brackish water (Fig. 4f). In general, the major ion/Cl ratios begin to approximate more closely the observed mixing lines as salinity increases resulting from other processes being overwhelmed by the total solute load.

Table 1 Hydrochemical data from surface water and groundwater, including the Yellow River and seawater

Group	Sample	Date	Temp (°C)	pH	EC ($\mu\text{S}/\text{cm}$)	ORP (mv)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Na^{+} (mg/L)	K^{+} (mg/L)	Br^{-} (mg/L)	HCO_3^{-} (mg/L)	Cl^{-} (mg/L)	SO_4^{2-} (mg/L)	NO_3^{-} (mg/L)	$\delta^{18}\text{O}$ (‰)	δD (‰)
Fresh	G7	2010/10/20	19.6	7.89	769	-21.1	22.6	10.0	68.1	3.9	5.5×10^{-1}	323.3	239.8	38.9	4.4	-7.6	-50
	G15	2010/10/20	15.7	7.38	2120	20.7	42.4	28.9	95.2	1.2	5.2×10^0	488.0	332.4	17.3	0.7	-9.0	-58
	G22	2010/10/22	17.4	7.54	409	-76.4	74.0	17.5	69.9	12.5	2.2×10^{-1}	195.2	81.4	33.1	0.9	-10.6	-70
	G8	2010/10/22	20.3	7.58	457	59.9	158.8	61.7	262.9	54.6	3.5×10^{-1}	228.8	235.0	131.5	4.8	-8.7	-58
	G10	2010/10/21	16.1	7.46	1123	-30.7	90.6	53.6	52.0	2.3		564.3	100.9	34.6	1.2	-9.3	-63
Brackish	G5	2010/10/20	17.1	7.43	12,660	-43.9	593.4	738.4	4796.4	64.0		564.3	10,382.8	1530.7	1.1	-5.4	-44
	G6	2010/10/20	19	7.38	2150	-71.5	296.6	358.0	2188.9	33.2	15.5×10^0	454.5	6176.5	221.3	10.9	-7.8	-56
	G9	2010/10/20	17.7	7.41	5560	-49.1	220.4	190.2	1520.1	10.1	16.4×10^0	1006.5	6365.6	109.9	1.4	-6.4	-48
	G11	2010/10/21	17.6	7.36	2580	-32.6	153.8	121.8	567.2	5.9	4.6×10^0	597.8	1844.2	160.3	1.0	-6.1	-45
	G16	2010/10/24	18.7	7.33	11,730	-249	518.0	590.8	3824.2	46.0		695.4	8357.9	1040.2	1.7	-5.5	-42
	G17	2010/10/23	16.8	7.29	4030	-155.1	193.6	193.9	1107.7	9.0	6.7×10^0	1091.9	3140.5	358.6	11.2	-6.0	-48
	G18	2010/10/25	18.9	7.43	1296	90.5	96.0	63.0	303.1	2.3	6.0×10^0	552.1	2495.3	291.4	10.7	-8.1	-57
	G19	2010/10/24	19.7	7.36	2610	-272.4	133.0	114.4	425.5	120.1	80.9×10^0	1037.0	1535.0	44.6	0.2	-6.8	-46
	G20	2010/10/24	18.4	7.17	3760	42.3	111.8	58.3	2775.6	16.8	20.9×10^0	823.5	8797.2	178.1	35.7	-8.8	-59
	G21	2010/10/20	19	7.66	872	136.4	113.6	53.0	223.3	6.6	6.3×10^{-1}	332.5	291.7	241.4	16.9	-4.6	-36
Salt	G23	2010/10/21	15.4	7.44	2040	91.9	148.2	81.2	176.0	1.6	5.1×10^{-1}	576.5	436.1	156.0	0.6	-8.1	-61
	G1	2010/10/21	22.4	7.05	11,200	-13.4	1087.0	958.0	4726.7	91.7	33.1×10^0	649.7	11,835.9	1252.3	0.7	-7.6	-59
	G2	2010/10/25	19.6	7.28	13,590	-64.2	903.4	907.7	4118.2	26.5	34.8×10^0	460.6	12,926.4	1242.7	3.8	-5.0	-37
	G3	2010/10/22	19.8	7.08	13,960	3.2	977.2	888.7	3767.4	35.5		445.3	12,415.7	1114.6	0.1	-4.4	-41
	G4	2010/10/24	19.7	8.48	23,300	-16.5	723.0	959.9	5098.9	42.9	41.7×10^0	762.5	17,376.5	1010.4	1.3	-5.9	-45
Yellow River	G12	2010/10/24	21.3	7.06	13,760	-25.7	588.4	650.4	3778.0	23.8	41.1×10^0	802.2	14,933.8	937.4	1.6	-5.6	-44
	G13	2010/10/22	18.2	7.09	14,090	-22	1050.0	967.9	4186.0	64.4	38.3×10^0	753.4	15,360.3	938.4	0.9	-3.6	-33
	G14	2010/10/24	20.8	7.82	22,800	9.2	910.4	1255.2	7912.7	117.4	68.3×10^0	686.3	25,113.5	2211.4	0.5	-3.5	-29
	YR1	2010/10/23	18.9	8.49	615	180.8	51.4	11.5	61.9	3.9		183.0	153.6	66.7	13.1	-9.1	-68
	YR2	2010/10/23	19.4	8.46	799	120	45.4	13.2	71.1	3.9		137.3	50.7	70.6	11.8	-6.4	-52
Seawater	SW	2010/10/22	18.5	8.41	15,460	185.9	773.8	805.6	5304.5	98.7	41.8×10^0	420.9	13,342.6	1595.0	7.8	-2.0	-30
Surface water	S1	2010/10/20	20.6	7.99	6990	182.7	113.8	164.9	1964.4	35.1	2.1×10^{-1}	305.0	4921.3	263.0	3.1	-5.1	-44
	S3	2010/10/20	23.2	8.71	14,550	194.9	87.2	84.6	345.0	5.9		146.4	879.8	251.5	2.5	-4.1	-34
	S4	2010/10/20	25	9.27	1744	158.1	53.6	35.3	458.2	18.7	4.8×10^{-2}	58.0	1045.0	71.0	1.1	-4.5	-39
	S5	2010/10/20	21.9	8.69	6270	229.9	204.8	186.6	2332.9	60.1	2.3×10^{-1}	122.0	4831.5	365.3	3.5	-6.2	-47
	S6	2010/10/21	21.9	8.5	12,680	174.7	169.4	347.0	4395.3	143.1	5.1×10^{-1}	161.7	10,646.1	668.2	0.6	-5.4	-43
	S7	2010/10/21	19.1	8.3	5520	205.5	17.4	15.4	138.2	2.3	5.6×10^{-2}	262.3	166.4	21.6	1.5	-5.6	-43
	S8	2010/10/21	21.7	7.8	5040	62.4	82.2	113.2	1331.7	21.5	1.3×10^{-1}	292.8	3636.9	177.6	1.2	-6.2	-52
	S9	2010/10/21	20.9	7.86	1140	14.5	37.2	32.5	290.3	7.4	8.0×10^{-3}	115.9	320.4	93.1	2.3	-4.3	-38
	S10	2010/10/22	22.5	8.2	1517	155.4	100.6	56.3	449.2	13.7	2.8×10^{-1}	137.3	1217.0	109.0	1.7	-6.3	-49

Table 1 continued

Group	Sample	Date	Temp (°C)	pH	EC ($\mu\text{S}/\text{cm}$)	ORP (mv)	Ca ²⁺ (mg/L)	Mg ²⁺ (mg/L)	Na ⁺ (mg/L)	K ⁺ (mg/L)	Br ⁻ (mg/L)	HCO ₃ ⁻ (mg/L)	Cl ⁻ (mg/L)	SO ₄ ²⁻ (mg/L)	NO ₃ ⁻ (mg/L)	$\delta^{18}\text{O}$ (‰)	δD (‰)
	S11	2010/10/22	21.8	7.92	1679	122	27.0	11.5	65.1	3.9		176.9	87.8	68.2	6.3	-6.5	-51
	S12	2010/10/20	17.5	7.64	3770	107.4	82.6	62.9	633.4	19.9	3.2×10^{-2}	277.6	1115.6	128.2	0.9	-9.6	-68
	S13	2010/10/22	19.5	8.15	15,190	116.5	162.8	214.8	2659.0	42.9		189.1	5492.6	193.9	5.3	-5.4	-45
	S14	2010/10/22	20.2	8.52	458	140.8	40.8	12.5	63.9	4.3		180.0	135.7	59.5	16.2	-4.7	-40
	S15	2010/10/23	18.6	8.46	5030	222.5	77.4	58.2	753.9	21.1	7.2×10^{-2}	193.4	1810.3	98.4	0.1	-7.1	-55
	S16	2010/10/23	19.4	8.34	497	220.8	53.6	19.7	96.4	17.6	8.0×10^{-3}	225.7	100.5	34.6	1.1	-8.1	-51
	S17	2010/10/23	17.5	7.99	1033	21.4	50.0	24.8	153.9	9.8		262.3	314.0	94.1	0.2	-5.1	-42
	S18	2010/10/24	20.2	8.23	9140	10.4	69.0	136.8	1828.7	38.2	1.1×10^{-1}	329.4	3629.3	281.3	23.9	-5.9	-48
	S19	2010/10/24	20	9.1	4060	48.4	36.8	67.0	813.3	28.1	4.8×10^{-2}	176.9	1011.1	205.4	56.2	-6.8	-54
	S20	2010/10/24	19.5	7.99	1635	127.8	135.4	32.2	306.1	11.7	1.6×10^{-2}	332.5	595.3	220.8	9.5	-4.3	-39
	S21	2010/10/24	19.7	8.31	2980	105.9	56.2	61.4	739.9	9.4	2.4×10^{-2}	253.2	1859.7	118.1	1.9	-1.6	-22
	S22	2010/10/25	18.9	8.16	2940	103.7	62.4	46.1	319.2	9.8	4.0×10^{-2}	338.6	634.8	97.9	0.4	-7.6	-58
	S23	2010/10/25	21.8	8.12	4010	154	97.0	103.9	1227.1	38.2	1.3×10^{-1}	247.1	2831.3	104.2	0.8	-5.8	-48

Saturation indices

Most water samples are at or above calcite and dolomite saturation, indicating the possibility of calcite and dolomite precipitation with increasing salinity (Fig. 5). The increase in the concentration of Ca²⁺ of saltwater compared to the mixing line thus can be explained by cation exchange rather than carbonate dissolution. The relative decrease in Ca²⁺ concentration in the freshwater and brackish water is thought to be due to some precipitation of carbonate minerals and reverse cation exchange. The SI for gypsum reaches saturation only in the saltwater, while the negative gypsum indices in freshwater and brackish water indicate the possibility of dissolution. The evolution of halite saturation indices shows an increasing trend plotted against the EC concentrations; all samples are below saturations (SI_{halite} < 0).

Stable isotopic composition

Stable oxygen and hydrogen isotopes could be indicators of groundwater sources, mixing process, and evaporation conditions (Li et al. 2008; Zhang et al. 2009) because they are generally considered to be transported conservatively in shallow aquifer settings. The local meteoric waterline (LMWL: $\delta\text{D} = 7.8 \times \delta^{18}\text{O} + 6.3$, $n = 8$, $r^2 = 0.78$ data from March to October) is determined by $\delta^{18}\text{O}$ and δD values of the average monthly rainfall monitored between 1986 and 1990 at the Yantai station which has similar climatic and coastal conditions (N37°32'24", E121°24'00"), about 260 km SE of DongYing City (IAEA/WMO 2006).

The groundwater reveals a very wide variation in the stable isotope content similar to the surface water, with $\delta^{18}\text{O}$ values ranged from -7 to -3 ‰ and δD values ranged from -73.4 to -28.9 ‰ (Table 1). The freshwater plotted along and between the LMWL and GMWL both depleted in $\delta^{18}\text{O}$ and δD ($\delta^{18}\text{O}$ from -9 to -7 ‰ and δD from -58.4 to -42.5 ‰). The saltwater is enriched in $\delta^{18}\text{O}$ and δD values of between -5 to -3 and -45.4 to -28.9 ‰. The brackish water shows a large range of variation (between -9 to -5 ‰ for $\delta^{18}\text{O}$ and between -60.9 to -35.7 ‰ for δD). They are located in an intermediate slightly offset from the LMWL and fall on the extension of a mixing trend defined by the saltwater.

The evaporation line ($\delta\text{D} = 5.5 \times \delta^{18}\text{O} - 14.49$, $R^2 = 0.93$, $n = 22$) can be drawn from the intersection of the Yellow River and majority of the surface water samples, which plotted above and to the right of them (Fig. 6). This means that groundwater probably contains a significant component of irrigation water which

Fig. 3 Piper plot for various waters in the YRD (**a**), and stiffmap plot of hydrochemical data for groundwater (**b**). Note that the groundwater was classed into three types: fresh (TDS <1 g/L), brackish (1–10 g/L), and salt (>10 g/L)

has undergone evaporation before infiltration. Direct evaporation from shallow water table is also expected to have occurred where water table transcends the critical water table depth (~ 3 m) concluded by Fan

(2010) and Wang et al. (2012) according to the field measurements, at which shallow groundwater is likely to be affected by continuous evaporation, causing precipitation of salts.

Fig. 4 Hydrochemical relationships between the concentration of Cl^- and $\text{Cl}^-/\text{HCO}_3^-$ (a), SO_4^{2-} (b), Na^+ (c), Na^+/Cl^- (d), Ca^{2+} (e), and Mg^{2+} (f); The line represents mixing between the two end-members: the fresh Yellow River and seawater. Numbers along the

mixing line show the percentage (%) of seawater with 10 % increments under simple mixing behavior. Concentration expressed in meq/L

Fig. 5 Saturation indices (SI) for halite, calcite, dolomite, and gypsum, all the various samples were calculated using PHREEQC version 2.8

Discussion

Origin and recharge of groundwater

The $\delta^{18}\text{O}$ and δD values of freshwater are mostly distributed close to the Yellow River ($\delta^{18}\text{O} \sim -9\text{‰}$, $\delta\text{D} \sim -67.4\text{‰}$) and slightly to the right of the LWML, indicating that the Yellow River and/or infiltration of local rainfall are their predominant origin. The brackish water shows much less influence from the local rainfall, since their $\delta^{18}\text{O}$ and δD values are within the range of evaporated surface waters.

The saltwater is characterized by both $\delta^{18}\text{O}$ and δD values plotted below the LWML and significantly lower than that of local seawater. The data do not support the fact that saltwater was formed due to simple evaporation-enrichment of seawater and was subsequently modified by mixing. Analysis of the conservative tracers of $\delta^{18}\text{O}$, δD , and Cl^- values in the YRD confined aquifer (>30 m depth) by Wang (2009) has shown the similar range with saltwater, indicating the same origin between the saltwater and the confined brine (Figs. 6, 7). Moreover, Chen et al. (2007) found there is a characteristic of upward gradient in the confined aquifer by comparing the water tables between the deep confined well (35 m depth) and shallow well (10 m) in the YRD coast. We propose the hypothesis that

the shallow saltwater at the coast stripe in the YRD was recharged by the upward flow from confined brines other than the intrusion of seawater. The other evidence ruling out the involvement of modern seawater is that the mean Cl/Br ratios of the saltwater ($\text{Cl}/\text{Br} = \sim 760$) are higher than the seawater ratio ($\text{Cl}/\text{Br} = \sim 639$). Chen et al. (2007) and Wang (2009) defined the confined brine (>30 m depth) derived from paleo seawater dated between 4000 and 7400 years BP using the ^{14}C dating different water origin. The fact that $\delta^{18}\text{O}$ and δD values of saltwater were much lower than that of seawater is likely explained by dilution of the paleo seawater with continental runoff prior or during the formation of brine, which is proposed by Han et al. (2014). This is consistent with the stable isotopic values of the local seawater (Fig. 6) which are isotopically depleted when compared to standard seawater ($\delta^{18}\text{O} \approx 0\text{‰}$ and $\delta\text{D} \approx 0\text{‰}$). The isotopic depletion may be an indication of large degree of dilution of water in the YRD with continental runoff.

Quantification of saline water intrusion

With increasing salinity, the freshwater and brackish water evolve toward more enriched $\delta^{18}\text{O}$ and δD ; while most brackish water shows a tendency to evolve toward saltwater which has mean $\delta^{18}\text{O}$ and δD of -5 and -41‰ ,

Fig. 6 Stable isotope data $\delta^{18}\text{O}$ versus δD values for all the water samples from surface water. The Yellow River, seawater, and groundwater in the YRD. Note that 1. **a** GMWL, the global meteoric waterline; **b** LMWL, the local meteoric waterline; **c** evaporation line. 2. Data from the literature in delta: confined aquifers in the coastline (depth >30 m) from Wang (2009)

Fig. 7 Relationship between Cl^- concentration and $\delta^{18}\text{O}$ values in groundwater, Yellow River water, and seawater. Symbols are the same as in previous figures

rather than local seawater which has mean $\delta^{18}\text{O}$ and δD of -2 and -30 ‰. This mixing trajectory between the Yellow River and saltwater is not obvious from the plot of ionic data alone as the seawater and saltwater have similar ionic ratios (Fig. 4).

The plot of Cl^- versus $\delta^{18}\text{O}$ can be used to further examine the mixing trajectory and quantities for two potential mixing lines, e.g., between the Yellow River and seawater or the Yellow River and saltwater (Fig. 7). Saturation with respect to halite is not found in any of the samples, excluding halite precipitation under current conditions (Fig. 5). Hence, Cl^- is used to behave relatively conservatively in the mixing model. Calculated by a mass-balance model, the mixing lines between two end-member samples were estimated and plotted based on:

$$C_m = C_f \times X + C_{\text{seawater/salt}} \times (1 - X).$$

where X is the mixing fraction of fresh end-member (the Yellow River), C_m is the $\delta^{18}\text{O}$ (‰) or Cl^- (meq/L) of the mixed groundwater; C_f is the mean $\delta^{18}\text{O}$ or Cl^- of the Yellow River; $C_{\text{seawater/salt}}$ for the $\delta^{18}\text{O}$ or Cl^- of seawater or saltwater (separate lines on each plot in Fig. 7).

The first obvious trend in this plot is that most brackish samples are distributed between the Yellow River (with Cl^- concentrations of ~ 1.08 meq/L and $\delta^{18}\text{O}$ value of ~ -9 ‰) and the saltwater (with Cl^- concentrations of ~ 629 meq/L and $\delta^{18}\text{O}$ value of ~ -3 ‰). The saltwater mixing proportion were estimated to increase from 5 % to 50 % of the mixed waters (freshwater and brackish water), with increasing proportion of saltwater from inland toward the coast. The saltwater mixing pattern has important implications for evaluating the extent of seawater intrusion; one unit volume mixing with saltwater causes much greater salinization contamination compared to that of local seawater. The second trend shows four brackish groundwater samples (G11, G17, G19, and G23) appear to fit the Yellow River–seawater mixing trajectory better than the Yellow River–saltwater mixing, suggesting classic seawater intrusion mechanism. Three of the four brackish water samples (G11, G19, and G23) are located near the coastline where the local surface water is more vulnerable to the seawater spray and then infiltrated into the aquifer. In the case of G17 located in the ancient riverbed of the Yellow River, it may be attributed to be a mixture of freshwater and fossil seawater trapped in the clay sediment (Ren and Shi 1986; Wolters et al. 2016). Apart from the most groundwater scattered close to the two trends, the high values of $\delta^{18}\text{O}$ observed in some freshwater and brackish water with relatively low Cl^- contents likely point to the local irrigation return flow evaporation prior to recharge.

Water–rock interaction

To further delineate the importance of cation exchange process, the concentration of Na^+ and Ca^{2+} was presented quantitatively by calculating the excess concentration (Na_{react} , Ca_{react}) as suggested by Appelo and Postma (2005) (Fig. 8). Except for G2, all the saltwater is depleted in sodium by -6 to -43 meq/L compared to the concentration expected from a simple binary mixing between the Yellow River and saltwater. However, most freshwater and brackish water samples show a slight freshening trend, with Ca^{2+} depletion (from -6.0 to -1.8 meq/L) observed together with Na^+ enrichment (0.8 to 12.5 meq/L). A comparison between the water levels (Fig. 2) and the salinization of the coastal aquifers shows this situation seems plausible in the YRD where the inland aquifer has been freshening by

Fig. 8 Ca_{react} and $\text{SO}_4_{\text{react}}$ versus Na_{react} plot for the groundwater. $\text{Me}_{\text{react}} = \text{Me}_{\text{measured}} - \text{Me}_{\text{mixing}}$, where $\text{Me}_{\text{mixing}}$ is the composition expected from a binary mixing between saltwater and the Yellow River, assuming the Cl^- is conservative

irrigation diverted from the Yellow River. The timescale of freshening process, which was determined by complicated factors such as the hydrological gradient, the porosity, and the cation exchange capacity of the sediment, may be on the order of decades to centuries. On the other hand, the process of freshening is expected to deplete- Ca^{2+} water in brackish groundwater. However, the freshwater and brackish water are largely near zero, with either slightly enriched or slightly depleted in Ca^{2+} (the mean Ca^{2+} deviation from mixing line is less than 3 meq/L, which is much lower than Na_{react}). The slight Ca^{2+} depletion related to freshening due to cation exchange may be blurred by the Ca^{2+} excess resulting from gypsum dissolution within the coastal aquifer ($\text{SI}_{\text{gypsum}} < 0$ in the freshwater and brackish water, Fig. 5). In contrast, it is notable that the reverse cation exchange process (Na^+ depletion and Ca^{2+} enrichment) appears significantly in the saltwater.

The sulfate depletion observed in the freshwater and brackish water (Fig. 4) has also been described in

association with freshwater flowing into coastal aquifer (Gattacceca et al. 2009). This may be attributed to reduction of SO_4^{2-} under anaerobic environment caused by fast vertical recharge in the irrigation zones (transformation of SO_4^{2-} into sulfide). This behavior is confirmed by the calculation of SO_4^{2-} deficiencies ($\text{SO}_{4\text{react}} < 0$, Fig. 8). This reaction needs organic material and SO_4^{2-} (Mollema et al. 2013), both of which are present in freshwater input from surface water in the YRD. Given that the SI of gypsum is quite negative in most groundwater samples, the depletion of SO_4^{2-} cannot be explained by gypsum precipitation, but is likely due to SO_4^{2-} reduction processes.

Conclusions

This study uses a combination of hydrodynamic, hydrochemical, stable isotopic measurements to explore groundwater salinization processes in the YRD, an example of complex groundwater table dynamics and salinization mechanisms in the reclaimed coastal plain of northern China. The evidence highlights the beneficial influence of the diverted water from the Yellow River which supplies large amounts of freshwater into aquifer system and confirms high degree of groundwater salinization is restricted only to the coastal strip.

Integration of different hydrochemical results (water types, major ionic ratios, and saturation indices) suggested more than one processes control the evolution in groundwater chemical compositions. First, the freshwater and brackish water, in the northern and central regions, represent the mixing of groundwater with irrigation and saline water enclosed in sediment; however, many other reactions can be triggered during a long residence time of groundwater due to irrigation recharge, jointly with an almost nil regional hydraulic gradient. Secondly, deviations from the mixing line for the major ions (Ca^{2+} , Na^+ , SO_4^{2-} , and Mg^{2+}) indicate several concurring and competing water-rock interactions, which also affect the salinization, such as cation exchange, calcite and gypsum dissolution/precipitation, halite solution, and SO_4^{2-} reduction. In the contrast, saltwater in the northern and eastern coastal lines reflects further mineralization, since the tidal flat without any vegetation makes the characteristic evapor-concentration possible in the semiarid climate.

The isotopic data ($\delta^{18}\text{O}$ and δD) are examined in conjunction with the Cl^- concentrations identifying the saltwater as saline end-member rather than seawater. The saltwater is recharged by the upward flow of saline confined aquifer, which is supported by the similar range of stable isotope ($\delta^{18}\text{O}$ and δD) and Cl^- values within them and the upward potential in the confined aquifer. Furthermore, relative depleted isotopic signatures of saltwater

compared to local seawater point to a former and initially depleted seawater reservoir resulting from runoff dilution in the saltwater. The saltwater, which can occur in coastal systems resulting from the history of sea-level transgression, regression, and evaporative enrichment, is likely to pose an equal or greater water quality threat than seawater, and to cause more serious soil degradation due to their higher salinity.

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