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Articles

Hydro-chemical characteristics and water environment of the surface water and shallow groundwater in the North of Yellow River Delta, China

Características hidroquímicas y ambiente acuático de las aguas superficiales y subterráneas poco profundas en el norte del delta del río Amarillo, China

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Abstract

The present study envisaged the dynamic monitoring and study of the hydrochemistry of the surface water and shallow groundwater in the

northern part of the Yellow River Delta, China. Analysis of the surface water and shallow groundwater revealed that the shallow groundwater exhibited in two forms: precipitation evaporation type and hydrological type, and the latter was obviously affected by the dynamic water level of the Yellow River. The surface water and shallow groundwater exhibited a higher degree of mineralization. Cations of Mg^{2+}, Na^{+} and K^{+} and anions of Cl^{-} and SO_4^{2-} were the main ion components of the total dissolved solids. The hydro-chemical type of shallow groundwater was Cl-Na·K that was a typical seawater or brine form. The water chemical type of surface water changed to $Cl·SO_4$-Na·K·Ca due to the industrial, agricultural and urban pollution. The overall quality of surface water was poor, and the main pollution indexes were found to be present, except for cyanide. The levels of COD and fluorine exceeded the standard values in the surface water. The groundwater quality was poor, since most of the pollutants were detected, except for cyanide and volatile phenol. Eight indexes exceeded the groundwater quality standard levels including Cl^{-}, Na^{+}, total hardness, COD, NH_4^{+}, Hg, As, NO_3^{-}, Cl^{-} and Na^{+}. The pollution of the surface water and shallow groundwater was mainly caused by the discharge of the wastewater from the petroleum; chemical industry and brine enterprises; agricultural non-point source pollution; leakage of septic tanks and

sewage pipes in urban and rural areas, and rainwater leaching from the garbage piles. In order to improve the water quality, the sewage control and management needs be optimised, and the industrial layout should be reconsidered.

Keywords: Surface water, shallow groundwater, dynamic type of groundwater, hydro-chemical characteristics, water pollution.

Resumen

El presente estudio contempló el seguimiento dinámico y el estudio de la hidroquímica del agua superficial y del agua subterránea poco profunda en la parte norte del delta del río Amarillo, China. El análisis del agua superficial y el agua subterránea poco profunda reveló que el agua subterránea poco profunda se exhibía en dos formas: tipo de evaporación por precipitación y tipo hidrológico, y este último fue obviamente afectado por el nivel dinámico del agua del río Amarillo. El agua superficial y el agua subterránea poco profunda exhibieron un mayor grado de mineralización. Los cationes de Mg^{2+}, Na^{+} y K^{+} y los aniones de Cl^{-} y SO_4^{2-} fueron los principales componentes iónicos del total de sólidos disueltos. El tipo hidroquímico de agua subterránea poco profunda era Cl-Na·K que era una forma típica de agua de mar o salmuera. El tipo químico del agua del agua superficial cambió a $Cl{\cdot}SO_4^{-}Na{\cdot}K{\cdot}Ca$ debido a la contaminación industrial, agrícola y urbana. La calidad general del agua superficial era mala y se encontró que estaban presentes los principales índices de contaminación, excepto el cianuro. Los niveles de DQO y flúor excedieron los valores estándar en el agua superficial. La calidad del agua subterránea fue mala, pues se detectaron la mayoría de los contaminantes, excepto cianuro y fenol volátil. Ocho índices excedieron los niveles estándar de calidad del agua subterránea, incluidos Cl^-, Na^+, dureza total, DQO, NH^{4+}, Hg, As, NO_3^-, Cl^- y Na^+. La contaminación de las aguas superficiales y subterráneas fue causada principalmente por la descarga de aguas residuales de la industria petrolera, química y de salmuera; contaminación de fuentes agrícolas difusas; fugas de fosas sépticas y tuberías de alcantarillado en áreas urbanas y rurales; y agua de lluvia que se mezcla con la lixiviación de las pilas de basura. Para mejorar la calidad del agua es necesario optimar el control y la gestión de las aguas residuales, y reconsiderar el diseño industrial.

Palabras clave: agua superficial, agua subterránea poco profunda, tipo dinámico de agua subterránea, características hidroquímicas, contaminación del agua.

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Introduction

The hydro-chemical characteristics of water are indicative of the climate and environment in the area, where the river flows. It is an important factor determining the use of water for domestic, irrigation, or industrial purposes, and plays a vital role in the sustainable management of the water resource utilization and the protection and construction of the ecological environment (Zhang, Ma, Abuduwaili, Ge, & Saparov, 2019).

The chemical composition of the water can be used to assess the water quality and evaluate its potential for different uses (Ren, Li, He, Su, & Elumalai, 2021; Zhan, Wu, & Jin, 2021). Groundwater water resource accounts to more than 50 % of the world's drinking water demand, and is essential for the daily life, industrial and agricultural production, and maintaining the ecological balance (Amiri, Bhattacharya, & Nakhaei, 2021a; Amiri, Kamrani, Ahmad, Bhattacharya, & Mansoori,2021b). However, groundwater pollution caused by the discharge from the industry (wastewater, waste gas, and solid waste); agriculture (excessive fertilizer application and pesticide spraying residues), and the local wastewater discharge affects the human health and reduces the agricultural food production (Zhang *et al*., 2019). Groundwater chemical composition is mainly affected by the natural (meteorology, surface water, plant soil, stratigraphic lithology) and anthropogenic (mixed groundwater extraction, pollutant discharge, artificial recharge) factors, and it is a product of long-term interactions with the surrounding environment (atmosphere, surface water, rocks) in the process of circulation (Iqla *et al*., 2019). Groundwater quality is jointly influenced by the water–rock action, human activities, and the atmosphere, which provide a basis for evaluating the water quality and scientifically formulating reasonable groundwater development and utilization plans (Liu *et al*., 2015).

The northern part of the Yellow River Delta is located in the south bank of the Bohai Sea and the north side of the Yellow River Estuary (He *et al*., 2017). The geomorphic type belongs to the typical alluvial plain area of the estuary Delta, which is from 37° 45′ ~ 38° 10′ north latitude to 118° 07′ ~ 119° 05′ east longitude (Figure 1).

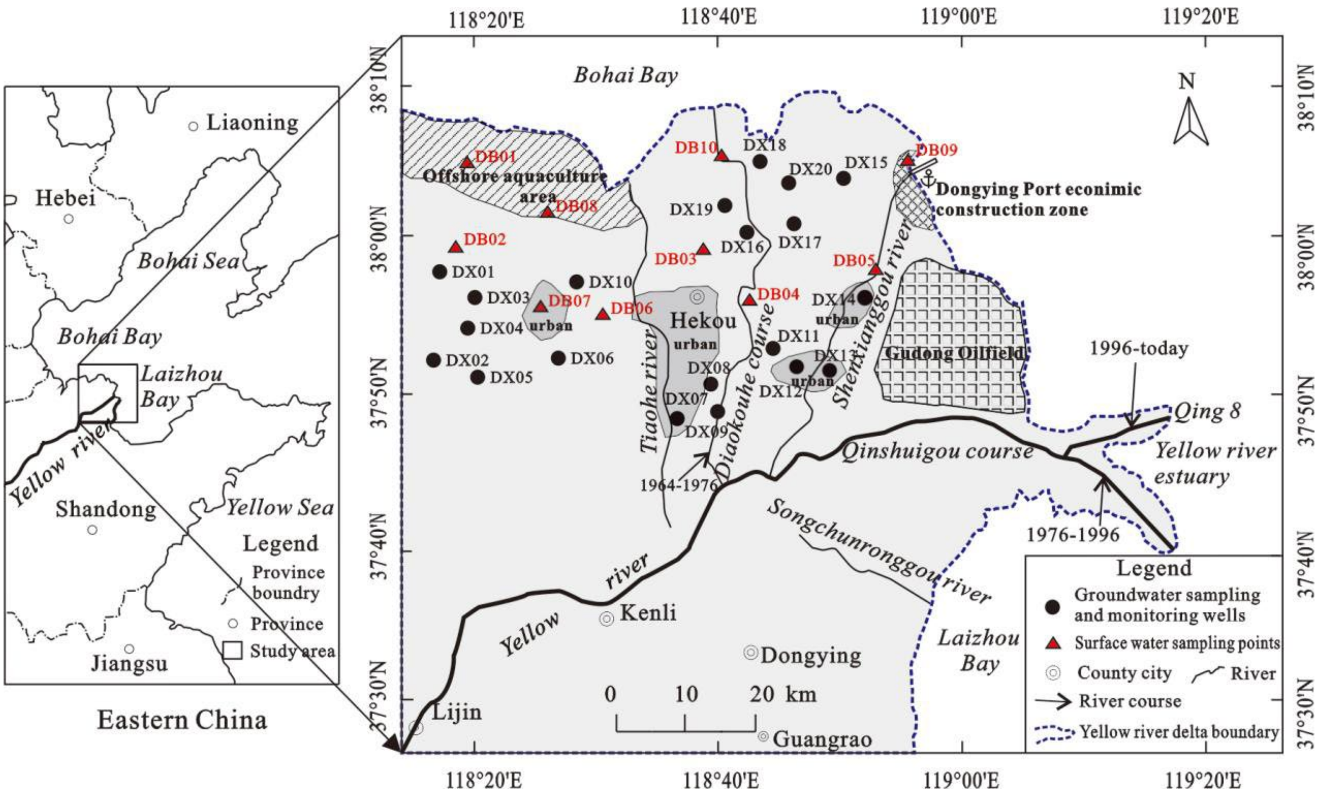

Figure 1. Spatial distribution of water sampling and monitoring sites.

Yellow River Delta, a typical transition area of the ocean, is one of the most important economic belts in China. Surface water and shallow groundwater are the dominant sources for the economic development in

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this area, and there is an increasing demand for water in the rapid economic development. However, the surface water and shallow groundwater are seriously affected by the intensified natural conditions, industry, agriculture and human activities. The surface water and shallow groundwater are characterized by high mineralization degree because of the adjacent ocean and the seawater intrusion. The total dissolved solids (TDS) levels of 0.8-20.0 g/l (the mean, 8.4 g/l) for surface water and 0.51-63.91 g/l for groundwater (Liu, Li, Li, Luo, & Huang, 2016); Ec of 1 010–33 200 μs/cm (Liu *et al*., 2014), and TDS of 0.8-32.89 g/l (the mean, 12.73 g/l) (Cao, Xu, Yu, & Huang, 2014) are documented. Additionally, the soil salinization also contributes to the high mineralization of the groundwater. At the same time, this area is the main location of Shengli Oilfield and salt industry, so some industries related to petrochemicals and salt engineering flourish in this region. The discharge of the landing oil and chemical enterprises enters into water. Besides the urban wastewater, urban solid waste and large-scale application of pesticides and fertilizers are reported to affect the water quality (Li, Xia, Zhao, & Chen, 2019; Wang, 2018; Wang, 2019; Wohlfart, Kuenzer, Chen & Liu, 2016; Zhao *et al*., 2018). Therefore, the process and mechanism governing the water characteristics and quality is complex in this area,

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and the study of hydro-chemical characteristics and water quality is essential for the sustainable development. However, the related research in this field is very limited. On the basis of the situations mentioned above, in the present study, the surface water and shallow groundwater were synthetically gathered in this area. The characteristics of surface water and shallow groundwater are discussed, and our results are expected to provide the scientific use of water, eco-environmental protection and the sustainable development.

Materials and methods

Study area

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Yellow River Delta is located in the north-western part of the Shandong province. The delta is comprised of the alluvial formed by the sediment in the Bohai depression. Quaternary unconsolidated sediments are widely distributed with a thickness of more than 400 meters. The sediment mainly originates from the silt carried by the Yellow River and the weathered rock (Wang & Qin, 2020). Additionally, there are three aquifers developed from the top to bottom in this area, *i.e.*, the unconfined and semi-confined aquifers (the 1st aquifer), confined middle porous aquifer (the 2nd aquifer), and the deep confined porous-fissure aquifers (the 3rd aquifer). The 1st aquifer is composed of fine sand, silt and clay silt during Holocene and late Pleistocene. The lower Pleistocene semi-confined aquifers are connected with the Holocene unconfined aquifer, and it belongs to a single shallow aquifer. The main recharge ways of the groundwater are atmospheric precipitation and river infiltration, and the discharge way is evaporation.

The area experiences three transgressions, and paleo-seawater (brine water) formed. The brine water flows through the ancient alluvial sand layer and intrudes into the coastal aquifers, thereby causing the seawater intrusion and soil salinization.

Sampling and analysis

Surface water sampling was collected in the main river channels (old channel of the Yellow River, Maxin River, Zhanli River, Tiaohe, etc.) in the north of the Yellow River Delta and near the estuary (Figure 1 and Figure 8). Totally 10 stations were involved for the study. Twenty groundwater monitoring wells were arranged for this study, and the wells are about 10m in depth (Figure 1). The groundwater levels were measured by a measuring clock three times a month, with a monitoring period of one year. The shallow groundwater was sampled from these monitoring wells from March to May 2017.

The surface water was taken by a plastic bottom. The groundwater was pumped into the collection barrel (Figure 2, Figure 3) and was filled into 1 000 ml plastic bottle. The bottle was washed for three times by the local water before sampling. Volatile phenols and cyanogens were sampled into 1 000 ml brown glass bottles separately, and NaOH was added to make pH > 12. The petroleum samples were sampled into 1 000 ml brown glass bottles separately and HCl was added to make pH < 2.

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The samples were sent to laboratory as soon as possible and analyzed in two weeks.

Figure 2. Groundwater sampling.

Figure 3. Surface water sampling.

The pH was analyzed with a PH-3C pH meter. COD was determined by employing 0.01 mol/l $KMnO_4$ in acid solution. TDS was determined by gravimetric method after drying the water sample at 105 °C. ICAP7400 inductively coupled plasma atomic emission spectrometry was used for the analysis of Ca^{2+}, Mg^{2+}, Na^+, K^+ and NH_4^+ was measured with a U-3310 spectrophotometer at a wavelength of 450 nm by reacting with potassium mercury iodide in an alkaline medium. Cl^- was determined by the titration method using 0.05 mol/l $AgNO_3$ with potassium chromate as indicator. F^- was measured by a fluoride selective electrode. SO_4^{2-} was determined by 0.01 mol/l EDTA titration with acid chrome blue k-naphthol green b indicator. Br-, after reaction with Chloramine T and Phenol Red, was measured by U-3310 spectrophotometer with a measurement wavelength of 590 nm. I^-, after reaction with bromine water, potassium iodide and starch in phosphoric acid medium, was measured by a U-3310 spectrophotometer with a measurement wavelength of 570 nm. HCO_3^- was determined by 0.01 mol/l HCl titration with phenolphthalein and helianthin B indicators. NO_3^- after reaction with ammonium sulphamate in hydrochloric acid medium, was measured by a U-3310 spectrophotometer with a measurement wavelength of 220 nm. ICAP RQ inductively coupled plasma mass spectrometry was used for the analysis of Cu, Pb, Zn, Mn, Cd, As, Se. Cyanide, after reaction with Chloramine T, Pyridine and Pyrazolinone, was measured by a U-3310 spectrophotometer with a measurement wavelength of 613 nm. Petroleum was measured by a U-3310 spectrophotometer at a wavelength of 256 nm after an extraction with petroleum ether. Hexavalent chromium, after reaction with Diphenylcarbazide in slightly acidic solution, was measured by a U-3310 spectrophotometer with a measurement wavelength of 540 nm. Volatile phenol was measured with a U-3310 spectrophotometer at a wavelength of 460 nm after a prior chloroform extraction.

For quality control, the blank, parallel and standard samples were also analyzed. The analysis tolerance was less than 5 %.

Analytical method

SPSS17.0 software was used to analyse the correlation of the geochemical properties. GW chart software was used to make Piper three-line diagram to analyze water ion type.

According to the environmental quality standard for surface water (GB3838-2002), the surface water quality in China is classified into I, II, III, IV and V, five categories. I represents a good water quality and V represents poor water quality. As the estuarine area was located in the offshore delta area, the overall surface water quality in the study area was relatively poor according to the background value of class V standard (Table 1). The water environment quality of the near sea area was evaluated according to the GB3097-1997 sea water quality standards and the background value of class IV standard (Table 2). The surface water quality was evaluated by the single factor evaluation method, which used the measured data and standard comparison classification, and selected the worst water quality category as the evaluation result. Therefore, the surface water detection indicators were compared according to the background value of class V standard of GB3838-2002 and GB3097-1997 environmental quality standard for surface water to see if they exceeded the standards. The indexes used included petroleum, volatile phenol, ammonia nitrogen (NH_4^+), fluorine (F^-), chemical oxygen demand (COD), selenium (Se), zinc (Zn), copper (Cu), arsenic (As), mercury (Hg), cadmium (Cd), lead (Pb), hexavalent chromium (Cr^{6+}), and cyanide (CN). According to the groundwater quality standard (GB/T14848-2017), the single factor evaluation method was used to evaluate the groundwater quality. Due to the high salt content and high salinity of the whole groundwater in the study area, the background value was based on class V (Table 3) for evaluation. The evaluation items included chloride, cyanide, volatile phenol, ammonia nitrogen, iron (Fe), chloride, sulfate, fluoride, iodide, nitrate, nitrite, total hardness, total dissolved solids (TDS), pH value, barium (Ba), selenium (Se), zinc (Zn), manganese (Mn), copper

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(Cu), cobalt (Co), nickel (Ni), molybdenum (Mo), arsenic (As), mercury (Hg), cadmium (Cd), lead (Pb), and hexavalent chromium (Cr^{6+}).

Table 1. Standard values of surface water, seawater and groundwater quality indexes.

Surface water quality, standard V value (mg/l)（GB3838-2002-class V）														
pH	COD	Cu	Zn	Se	As	Hg	Cd	Cr (Cr^{6+})	Pb	Cyanide	Volatile	Petroleum	F^-	NH_4
6~9	≦40	≦1.0	≦2.0	≦0.02	≦0.1	≦0.001	≦0.01	≦0.1	≦0.1	≦0.2	≦0.1	≦1.0	≦1.5	≦2.0
Seawater quality Standard Ⅳ(mg/l)（GB3097-1997 - class Ⅳ）														
pH	COD	Cu	Zn	Se	As	Hg	Cd	Cr (Cr^{6+})	Pb	Cyanide	Volatile	Petroleum	Ni	
6.8~8.8	≦5	≦0.050	≦0.50	≦0.050	≦0.050	≦0.0005	≦0.010	≦0.050	≦0.050	≦0.20	≦0.050	≦0.50	≦0.050	
Groundwater quality, standard V value (mg/l)（GB/T14848-2017-classⅤ）														
pH	Total hardness	Cl-	TFe	Mn	Cu	Zn	Al	Volatile	COD	NH_4	Na	Cyanide	Hg	As
<5.5 or >9.0	>650	>350	>2.0	>1.50	>1.50	>5.00	>0.50	>0.01	>10.0	>1.50	>400	>0.1	>0.002	>0.05
Se	Cd	Cr (Cr^{6+})	Pb	Be	Ba	Ni	Co	Mo	NO_3^-					
>0.1	>0.01	>0.10	>0.10	>0.06	>4.00	>0.10	>0.10	>0.15	>30.0					

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Table 2. Analysis of surface water samples.

Number	Quality grade	pH	COD	Cu (mg/l)	Zn (mg/l)	Se (μg/l)	As (μg/l)	Hg (μg/l)	Cd (μg/l)	Cr(Cr^{6+}) (μg/l)	Pb (μg/l)	Cyanide (mg/l)	Volatilized Phenol (mg/l)	Petroleum (mg/l)	F^- (mg/l)	NH_4^+ (mg/l)	Ni (mg/l)
DBY01	**Ⅳ**	8.02	2.18	<0.05	<0.002	<0.05	<10	<0.25	<1	<10	<10	/	0.036	0.05	0.99	<0.04	<0.005
DBY02	**V**	7.76	8.38	<0.05	<0.002	<0.05	<10	<0.25	<1	<10	<10	/	0.0362	0.06	0.63	0.08	<0.005
DBY03	**Ⅳ**	6.95	12.52	<0.05	<0.002	0.24	<10	<0.25	<1	<10	<10	/	/	0.01	1.11	0.04	<0.005
DBY04	**Ⅳ**	8.07	4.00	<0.05	<0.002	<0.05	<10	<0.25	<1	<10	<10	/	0.008	/	0.68	<0.04	<0.005
DBY05	**Inferior V**	8.05	8.05	<0.05	<0.0005	<0.05	<10	<0.25	<1	<10	<10	/	/	0.04	**1.92**	0.30	<0.005
DBY06	**Ⅳ**	7.83	4.46	<0.05	<0.002	<0.05	<10	<0.25	<1	<10	<10	/	/	0.03	0.63	<0.04	<0.005
DBY07	**V**	7.66	13.39	<0.05	<0.002	<0.05	<10	<0.25	<1	<10	<10	/	/	/	0.53	1.90	<0.005
DBY08	**Inferior Ⅳ**	7.12	**6.86**	<0.05	<0.002	0.1623	<10	<0.25	<1	<10	<10	/	0.0045	/	**19.35**	<0.04	<0.005
DBY09	**Ⅳ**	7.90	1.31	<0.05	<0.0005	<0.05	<10	<0.25	<1	<10	<10	/	/	/	1.02	<0.04	<0.005
DBY10	**Ⅳ**	8.04	22.42	<0.05	<0.0005	<0.05	<10	<0.25	<1	<10	<10	/	/	/	1.10	0.28	<0.005

Number	K^+ (mg/l)	Na^+ (mg/l)	Ca^{2+} (mg/l)	Mg^{2+} (mg/l)	TFe (mg/l)	Al^{3+} (mg/l)	Cl^- (mg/l)	SO_4^{2-} (mg/l)	HCO_3^- (mg/l)	CO_3^{2-} (mg/l)	I^- (mg/l)	Br^-(mg/l)	NO_3^- (mg/l)	NO_2^- (mg/l)	PO_4^{3-} (mg/l)	Ba (mg/l)
DBY01	346.50	10490.00	435.80	1337.00	<0.04	<0.04	19040.22	2670.38	155.96	9.59	0.12	56	15.55	0.44	<0.04	0.0566
DBY02	34.68	2256.00	190.20	364.40	<0.04	<0.04	4109.40	562.43	266.43	/	0.12	9.5	5.45	1.5	0.24	0.0939
DBY03	413.30	11920.00	579.20	1708.00	<0.04	<0.04	22190.76	3308.43	149.46	/	0.12	44.00	14.94	0.1	0.22	0.185
DBY04	6.61	513.10	107.10	102.00	<0.04	<0.04	938.31	330.84	188.45	/	<0.05	6.00	1.04	0.036	<0.04	0.2444
DBY05	383.20	12190.00	566.20	1719.00	<0.04	<0.04	21745.58	2629.02	103.97	44.74	0.1	45.00	18.43	<0.002	0.65	0.21
DBY06	18.24	1330.00	149.80	221.50	<0.04	<0.04	2362.91	425.37	259.93	3.20	0.06	8.00	15.17	0.50	<0.04	0.139
DBY07	107.30	5786.00	240.00	871.80	<0.04	<0.04	10889.91	1323.37	321.67	/	0.25	36.00	12.51	<0.002	0.32	0.1699
DBY08	404.00	11070.00	501.40	1451.00	<0.04	<0.04	19862.10	2883.06	136.46	/	0.15	64.0	16.61	0.20	0.22	0.1298
DBY09	376.40	10510.00	409.00	1142.00	0.057	<0.04	17567.68	2380.89	159.21	3.19	<0.05	47.5	18.28	0.18	<0.04	0.06
DBY10	120.20	4275.00	296.10	560.00	0.05	<0.04	7396.92	1157.95	120.22	12.78	<0.05	35.00	11.53	<0.002	<0.04	0.24

Number	Li(mg/l)	Sr (mg/l)	Mn (mg/l)	Co (mg/l)	Mo (mg/l)	Be (mg/l)	Sb (μg/l)	Total hardness (mg/l)	Permanent hardness (mg/l)	Temporary hardness (mg/l)	Total alkalinity (mg/l)	H_2SiO_3 (mg/l)	Free CO_2 (mg/l)	HBO_2 (mg/l)	TDS(mg/l)	Salinity (mg/l)
DBY01	0.1709	7.27	<0.01	<0.05	0.0059	<0.005	<0.5	6594.05	6450.14	143.91	143.91	1.402	/	15.31	34481.53	34559.51
DBY02	0.0519	3.53	0.094	<0.005	0.0097	<0.005	<0.5	1975.58	1757.06	218.51	218.51	0.8729	4.74	2.739	7693.40	7801.61
DBY03	0.2424	9.44	1.014	<0.005	0.0076	<0.005	<0.5	8479.93	8357.35	122.58	122.58	2.494	9.47	18.82	40256.75	40331.48
DBY04	0.0347	1.97	<0.01	<0.005	0.0075	<0.005	<0.5	687.49	532.94	154.56	154.56	0.5144	/	1.5	2100.34	2194.56
DBY05	0.25	10.85	0.14	<0.005	0.018	<0.005	/	8492.76	8332.87	159.89	159.89	10.42	/	15.58	39359.14	39411.13
DBY06	0.0402	2.71	1.06	<0.005	0.0076	<0.005	<0.5	1286.23	1067.71	218.52	218.52	1.73	/	1.852	4666.82	4796.58
DBY07	0.0774	4.33	0.9611	<0.005	0.0131	<0.005	<0.5	4189.40	3925.58	263.82	263.82	0.889	3.16	3.704	19431.16	19591.99
DBY08	0.2157	7.92	0.4796	<0.05	0.0254	<0.005	<0.5	7227.32	7115.40	111.92	111.92	4.871	7.89	17.99	36343.85	36412.08
DBY09	0.18	7.24	0.012	<0.005	0.021	<0.005	/	5724.12	5588.22	135.90	135.90	7.82	/	15.87	32541.76	32621.37
DBY10	0.14	7.23	0.012	<0.005	0.018	<0.005	/	3045.52	2925.60	119.91	119.91	7.6	/	7.48	13932.82	13992.93

DBY01, DBY08, DBY09 samples are sea water, figures in bold and with background.

Color are out of standard items.

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Table 3. Analysis of groundwater samples.

Number	pH	Total hardness (mg/l)	Cl^- (mg/l)	TFe (mg/l)	Mn (mg/l)	Cu (mg/l)	Zn (mg/l)	Al^{3+} (mg/l)	Volatile phenol (mg/l)	COD	NH_4^+ (mg/l)	Na^+ (mg/l)	Cyanide (mg/l)	Hg (μg/l)	As (μg/l)	Se (mg/l)
DXY01	6.84	5743.78	7670.88	<0.04	7.11	<0.05	<0.002	<0.04	/	2.39	1.00	2731.00	/	<0.25	<10	<0.05
DXY02	6.86	9777.2	29587.68	<0.04	1.23	<0.05	<0.002	<0.04	/	5.44	<0.04	16330.00	/	<0.25	<10	<0.05
DXY03	6.98	2907.09	2397.15	<0.04	2.55	<0.05	<0.002	<0.04	/	3.70	0.20	1080.00	/	<0.25	<10	<0.05
DXY04	6.83	10420.72	25615.26	<0.04	3.88	<0.05	<0.002	<0.04	/	4.35	<0.04	13110.00	/	<0.25	<10	<0.05
DXY05	7.04	11748.11	38354.4	<0.04	3.29	<0.05	<0.002	<0.04	/	6.31	<0.04	20290.00	/	<0.25	<10	<0.05
DXY06	7.11	10513.69	26574.12	<0.04	1.71	<0.05	<0.002	<0.04	/	5.12	<0.04	14150.00	/	<0.25	<10	<0.05
DXY07	6.76	8320.52	11711.79	<0.04	12.04	<0.05	<0.002	<0.04	/	4.24	0.16	4478.00	/	<0.25	<10	<0.05
DXY08	7.33	1152.45	1198.58	<0.04	1.52	<0.05	<0.002	<0.04	/	9.47	0.24	527.90	/	<0.25	<10	<0.05
DXY09	7.1	5902.01	9588.6	<0.04	6.38	<0.05	<0.002	<0.04	/	3.59	1.50	4521.00	/	<0.25	<10	<0.05
DXY10	6.98	15896.1	49315.07	<0.04	0.93	<0.05	<0.002	<0.04	/	4.35	<0.04	26980.00	/	<0.1	<10	<0.05
DXY11	7.06	7005.78	15615.72	<0.04	6.29	<0.05	<0.002	<0.04	/	3.05	0.36	7777.00	/	<0.25	<10	<0.05
DXY12	6.84	7439.74	17122.5	<0.04	7.68	<0.05	<0.002	<0.04	/	3.70	0.64	8956.00	/	<0.25	<10	<0.05
DXY13	7.6	580.99	1027.35	<0.04	0.61	<0.05	<0.002	<0.04	/	4.00	1.80	657.70	/	<0.25	<10	<0.05
DXY14	7.53	2099.69	3219.18	<0.04	2.37	<0.05	<0.002	<0.04	/	7.73	3.40	1466.00	/	<0.25	<10	<0.05
DXY15	6.93	4114.43	5171.00	0.06	5.02	<0.05	<0.0005	<0.04	/	3.59	3.40	2206.00	/	<0.25	<10	<0.05
DXY16	7.43	1436.25	1806.42	0.13	1.58	<0.05	<0.0005	<0.04	/	14.69	7.50	881.70	/	<0.25	<10	<0.05
DXY17	7.41	8653.35	17841.64	0.23	4.04	<0.05	<0.0005	<0.04	/	11.10	38.00	9301.00	/	<0.25	<10	<0.05
DXY18	10.5	2896.33	7966.04	<0.04	<0.01	<0.04	<0.009	<0.05	/	41.49	1.53	4543.50	/	4.03	44.05	<0.05
DXY19	6.92	11250.22	25208.54	0.14	6.27	<0.04	0.03	<0.06	/	10.81	1.06	13060.00	/	7.16	59.65	<0.05
DXY20	6.88	4316.57	8862.64	0.39	7.06	<0.04	0.14	<0.07	/	10.61	0.44	4593.50	/	2.04	19.00	<0.05

Number	Cd (μg/l)	Cr (Cr^{6+}) (μg/l)	Pb (μg/l)	Be (mg/l)	Ba (mg/l)	Ni (mg/l)	Co (mg/l)	Mo (mg/l)	NO_3^- (mg/l)	K^+ (mg/l)	Ca^{2+} (mg/l)	Mg^{2+} (mg/l)	SO_4^{2-} (mg/l)	HCO_3^- (mg/l)	F^- (mg/l)	I^- (mg/l)
DXY01	<1	<10	<10	<0.005	0.34	<0.005	<0.005	<0.005	11.90	16.22	1150	697.4	378.11	422.39	0.29	1
DXY02	<1	<10	<10	<0.005	0.05	<0.005	<0.005	0.01	36.51	458.6	648.5	1981	3710.17	562.11	0.25	0.9
DXY03	<1	<10	<10	<0.005	0.11	<0.005	<0.005	<0.005	14.56	16.45	474	418.5	850.74	929.26	0.3	0.19
DXY04	<1	<10	<10	<0.005	0.14	<0.005	<0.05	0.01	30.51	257	883.1	1995	2788.53	623.84	0.29	0.96
DXY05	<1	<10	<10	<0.005	0.11	<0.005	<0.005	0.01	49.42	566	507.7	2545	3851.96	649.83	0.35	0.6
DXY06	<1	<10	<10	<0.005	0.07	<0.005	<0.005	<0.005	42.66	324.1	407.5	2306	3048.48	770.05	0.53	2.4
DXY07	<1	<10	<10	<0.005	0.21	<0.005	<0.005	<0.005	24.51	17.75	1574	1066	1087.06	513.37	0.3	0.35
DXY08	<1	<10	<10	<0.005	0.35	<0.005	<0.005	<0.005	8.33	15.69	204.9	155.6	198.51	389.9	0.63	0.07
DXY09	<1	<10	<10	<0.005	0.35	<0.005	<0.05	0.01	19.12	22.58	753.3	976.4	1063.42	909.77	0.35	1.9
DXY10	<1	<10	<10	<0.005	0.06	<0.005	<0.005	<0.005	42.66	761.8	633.5	3476	5671.59	415.89	0.28	0.07
DXY11	<1	<10	<10	<0.005	0.14	<0.005	<0.005	<0.005	40.61	80.49	632	1318	1725.11	480.88	0.35	1.2
DXY12	<1	<10	<10	<0.005	0.16	<0.005	<0.005	0.01	58.38	98.07	756.3	1348	2150.48	747.31	0.33	2.4
DXY13	<1	<10	<10	<0.005	0.25	<0.005	<0.005	<0.005	10.99	16.91	79.21	93.05	94.53	539.36	0.59	0.34
DXY14	<1	<10	<10	<0.005	0.38	<0.005	<0.005	0.01	43.04	16.16	283.1	338.2	115.8	532.86	0.44	1.4
DXY15	<1	<10	<10	<0.005	0.1	<0.005	<0.005	<0.005	30.08	24.5	664.6	596.10	673.5	653.08	0.32	0.60

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DXY16	<1	<10	<10	<0.005	0.12	<0.005	<0.005	0.01	**42.02**	10.78	266.6	187.10	342.66	298.92	0.44	0.05
DXY17	<1	<10	<10	<0.005	0.1	<0.005	<0.005	<0.005	**149.73**	126.6	559.6	1762.00	1920.02	591.35	0.44	2.75
DXY18	<1	<10	<10	<0.005	0.17	<0.005	<0.005	0.04	**34.90**	139.75	1117.5	51.84	1317.61	/	0.67	0.08
DXY19	<1	<10	<10	<0.005	0.45	<0.005	<0.005	0.01	**39.74**	219.85	1252	2087.00	3172.02	474.38	0.40	0.25
DXY20	<1	<10	<10	<0.005	0.13	<0.005	<0.005	<0.005	**36.26**	67.92	615.85	646.90	988.21	601.1	0.43	0.35
Number	**Br^- (mg/l)**	**NO_2^- (mg/l)**	**PO_4^{3-} (mg/l)**	**Petroleum (mg/l)**	**Permanent hardness (mg/l)**	**Temporary hardness (mg/l)**	**Total alkalinity (mg/l)**	**H_2SiO_3 (mg/l)**	**Free CO_2 (mg/l)**	**HBO_2 (mg/l)**	**TDS (mg/l)**	**Salinity (mg/l)**	**Sb (μg/l)**	**Li (mg/l)**	**Sr (mg/l)**	**Quality grade**
DXY01	22	<0.002	<0.04	0.33	5397.35	346.43	346.43	17.4	29.99	0.83	12904.39	13115.58	<0.5	0.08	16.15	**Inferior V**
DXY02	100	0.01	<0.04	0.11	9316.18	461.02	461.02	13.66	34.73	7.33	53145.19	53426.24	<0.5	0.13	10.19	**Inferior V**
DXY03	5	1	<0.04	/	2144.96	762.14	762.14	17.6	45.78	1.51	5736.26	6200.89	<0.5	0.1	7.19	**Inferior V**
DXY04	72	<0.002	<0.04	0.01	9909.07	511.65	511.65	17.03	47.36	5.4	45077.67	45389.59	<0.5	0.14	14.85	**Inferior V**
DXY05	140	<0.002	<0.04	/		11748.11	532.96	13.1	25.26	8.62	66640.43	66965.34	<0.5	0.2	9.44	**Inferior V**
DXY06	70	<0.002	<0.04	/	9882.13	631.56	631.56	11.34	25.26	10.93	47319.54	47704.56	<0.5	0.17	8.26	**Inferior V**
DXY07	36	0.02	<0.04	1.19	7899.48	421.04	421.04	21.14	45.78	1.59	20268.90	20525.58	<0.5	0.16	21.07	**Inferior V**
DXY08	3.2	<0.002	<0.04	/	832.68	319.78	319.78	14.06	14.21	1.99	2519.42	2714.37	<0.5	0.04	3.14	**Inferior V**
DXY09	26	0.3	<0.04	0.02	31246.38	359.23	359.23	19.84	64.72	2.69	17444.62	17899.5	<0.5	0.11	12.96	**Inferior V**
DXY10	170	0	<0.04	0.01	15555.01	341.09	341.09	10.52	25.26	11.35	87267.02	87474.96	<0.5	0.26	11.06	**Inferior V**
DXY11	40	0.04	<0.04	/	15618.36	384.53	384.53	13.09	23.68	3.98	27481.39	27721.83	<0.5	0.11	10.67	**Inferior V**
DXY12	48	<0.002	<0.04	/	17604.01	373.05	373.05	21.71	63.15	6.81	30931.46	31305.11	<0.5	0.16	12.06	**Inferior V**
DXY13	2.2	<0.002	<0.04	/	138.63	442.36	442.36	20.05	6.31	2.11	2269.78	2539.46	<0.5	0.02	1.50	**Inferior V**
DXY14	8	0	<0.04	0.47	1662.66	437.03	437.03	15.05	11.05	2.53	5772.73	6039.16	<0.5	0.04	5.13	**Inferior V**
DXY15	12.5	<0.002	<0.04	/	3578.81	535.63	535.63	20.64	48.94	2.52	9725.02	10051.56	/	<0.02	10.80	**Inferior V**
DXY16	2.5	<0.002	<0.04	/	1191.09	245.16	245.16	22.36	9.47	0.61	3714.43	3863.89	/	0.05	4.06	**Inferior V**
DXY17	37.5	<0.002	<0.04	/	8168.35	485.00	485	36.68	25.26	6.8	32063.18	32358.85	/	0.11	11.23	**Inferior V**
DXY18	9.04	<0.002	<0.04	0.13	/	/	/	16.28	无	0.59	15100.00	15100.00	/	0.21	7.16	**Inferior V**
DXY19	87.96	0.13	<0.04	0.12	10861.15	389.07	389.07	16.54	20.52	2.33	45120.00	45357.19	/	0.27	20.76	**Inferior V**
DXY20	23.96	0.07	<0.04	0.09	3823.57	493.00	493.00	12.47	28.42	1.23	14820.00	15120.55	/	0.13	9.10	**Inferior V**

Figures in bold and with background colour are out of standard items.

Results and discussion

Characteristics of groundwater levels and the dynamic types

From the results of groundwater monitoring level elevations in 2017, the dynamic types of shallow groundwater level in this area can be divided into two types: Precipitation infiltration evaporation type and hydrological type (Figure 4).

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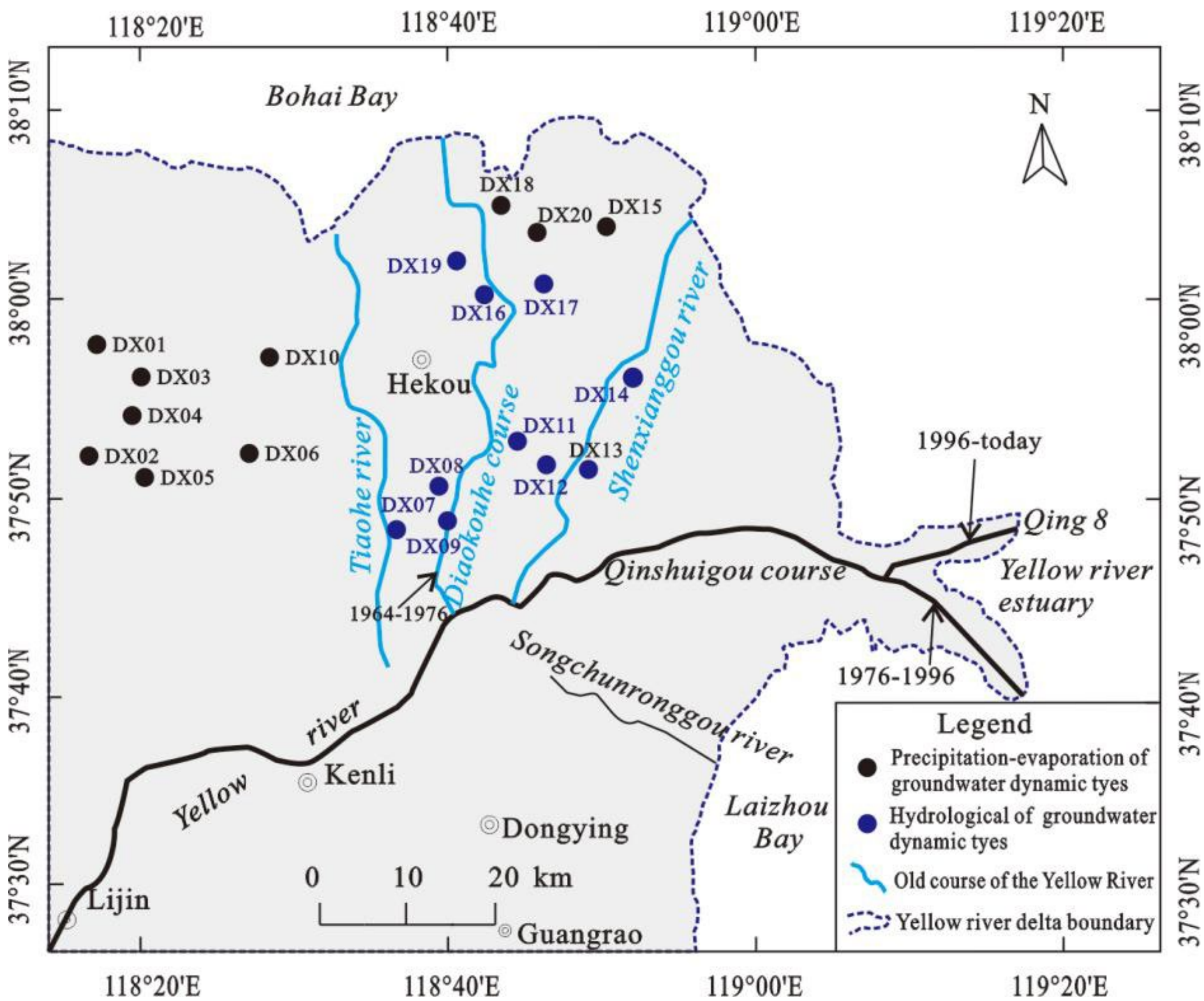

Figure 4. Distribution of monitoring wells of different groundwater dynamic types.

Precipitation-evaporation type: The groundwater level elevations are characterized by the groundwater cycle of single peak changing from flat, falling, rising to flat bottom. The precipitation was rare from April to

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May, and the groundwater entered into the dry season from May to June with the lowest levels. The groundwater level showed a rise and began to enter into the wet season, until the rainy season of July, and the peak value generally appeared in August. Such facts were consistent with the meteorological precipitation. After September, the precipitation became rare again, and the evaporation on the water surface also decreased correspondingly. The groundwater entered into the normal period; typical examples of DX01 and DX10 stations are shown in Figure 5. This groundwater mainly distributed in the coastal area and was mixed by brine or salt water. The main alternative modes were found to be precipitation infiltration and phreatic water evaporation.

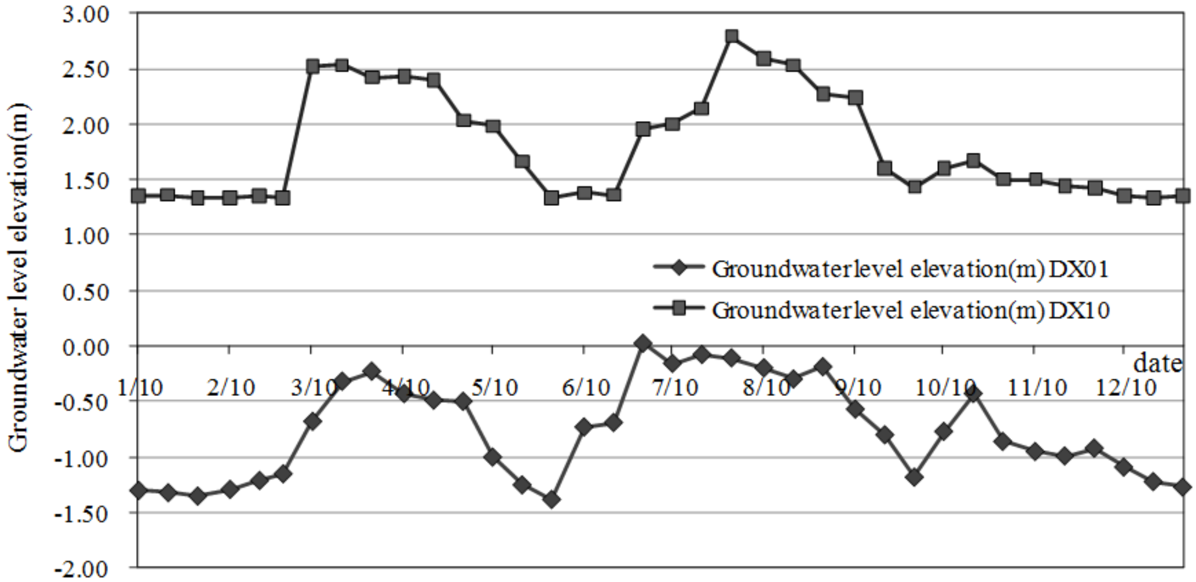

Figure 5. Example (DX01 and DX10 station) of dynamic curve of precipitation evaporation shallow groundwater level elevation Groundwater level elevation (m) = monitoring point elevation (m) - groundwater depth (m).

Hydrological type: The groundwater levels were characterized by double peaks and single bottom and multi peaks and multi bottom. Typical examples of DX11and DX16 stations are presented in Figure 6. This water was mainly distributed along the old Yellow River. The groundwater belonged to the shallow water and its dynamic state was not only affected by meteorological and irrigation infiltration factors, but also by the water level of the Yellow River. The closer the distance to the Yellow River, the more obvious was the hydrological impact of the Yellow River.

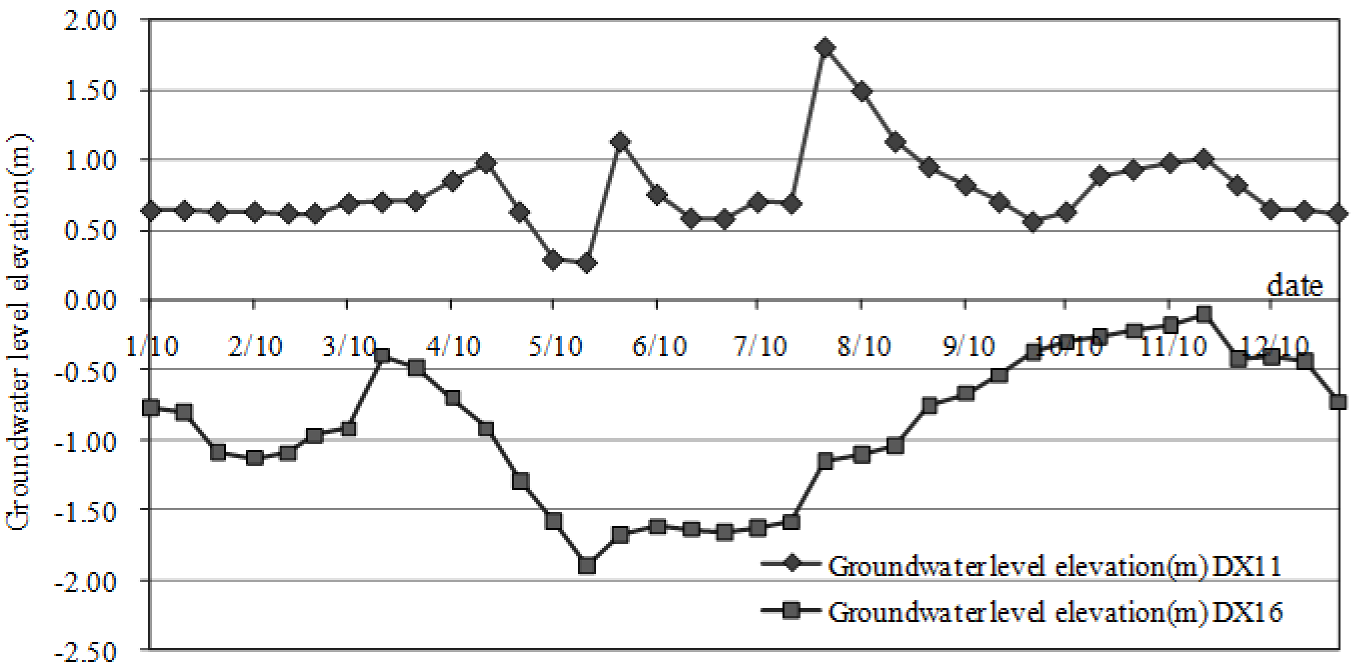

Figure 6. Example (DX11 and DX16 station) of dynamic curve of hydrological shallow groundwater level elevation

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Hydro-chemical characteristics of surface water and shallow groundwater

The Piper diagram is an accessible technique used in the hydro-chemical analysis, and is widely used to demonstrate the general chemical characteristics. The Piper diagram of the surface water and shallow groundwater in this area is illustrated in Figure 7. The main ions of the shallow groundwater were Na^+, K^+ and Cl^- (Figure 6). The hydro-chemical types were mainly Cl-Na and Cl-Na·K, which denote a typical mixed seawater or brine. The main ionic components of surface water were Na^+, K^+ and Cl^-, but due to the pollution from the irrigation and urban industry, the Ca^{2+}, Mg^{2+} and SO_4^{2-} levels increased, and the hydro-chemical types changed to Cl-Na·K and Cl·SO_4-Na·K·Ca, which made the hydro-chemical types more complex.

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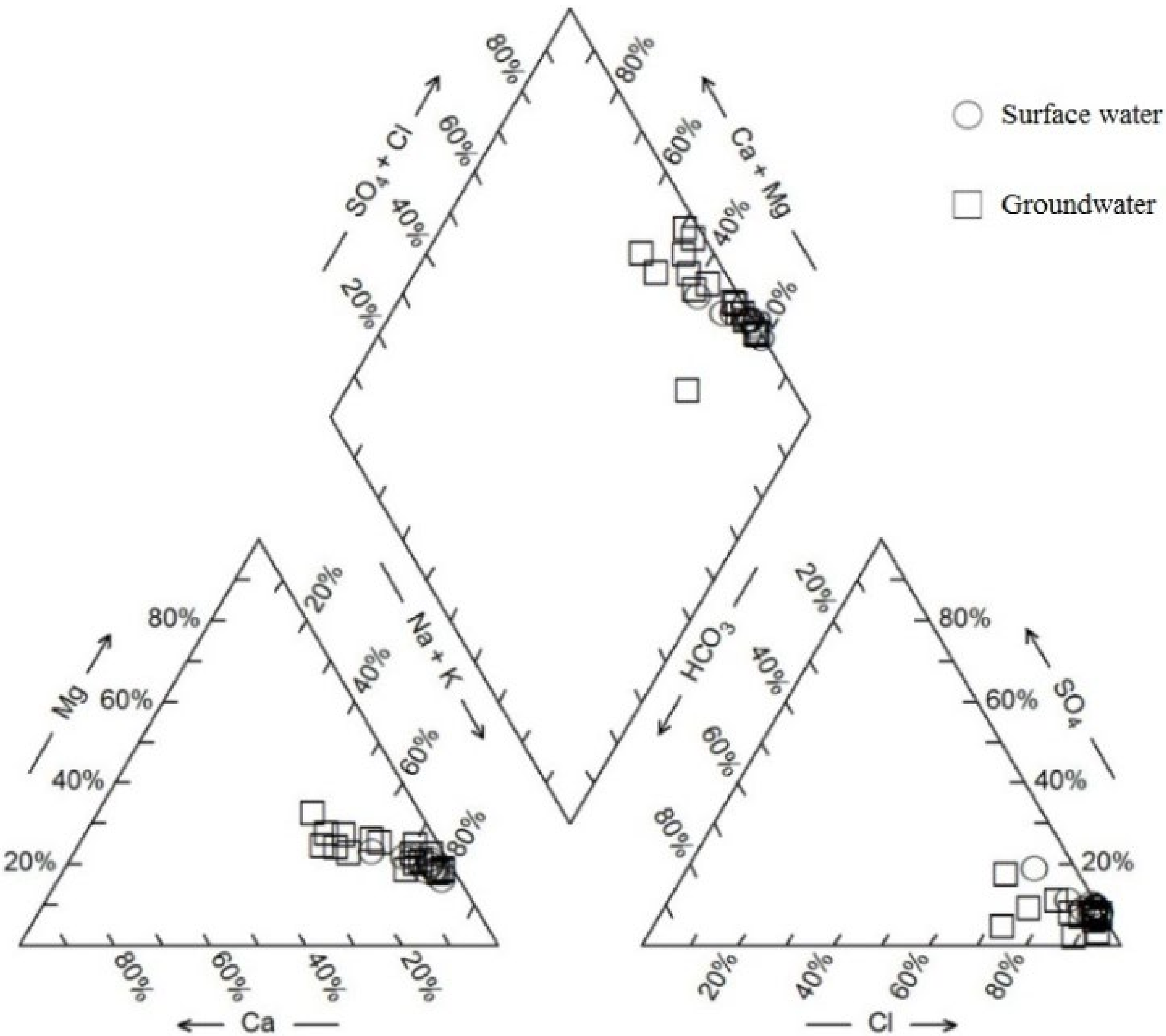

Figure 7. Piper graph of the surface water and shallow groundwater in the coastal region.

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The China Geological Survey has set the seawater intrusion standards of Cl^- 250 mg/l, Br- 0.55 mg/l, and TDS 1.0 mg/l. In the study area, the surface water consisted of 938-22 190 mg/l of Cl^-, 6-64 mg/l of Br^-, and 2 100-40 256 mg/l of TDS, and the shallow groundwater had 1 027-49 315 mg/l of Cl^-, 2.5-170 mg/l of Br^-, and 2 269-87 267 mg/l of TDS (Table 2). The surface water and shallow groundwater was typically characterized by salt water or brine water. Moreover, the TDS was closely related to the distance from the coastal line. The shorter the distance that the surface water monitoring point from the coastline is, the larger is the TDS is, while the shallow groundwater shows opposite characteristics. This means modern seawater significantly influenced the surface water, and the paleo-seawater in sediment influenced the shallow groundwater.

The correlation between TDS and main salts can be seen from the water type, and further analysis of the correlation can be seen from the correlation matrix in Table 4. It can be seen that the cations of Ca^{2+}, Mg^{2+}, Na^+ and K^+ in surface water exhibited a significant positive correlation with the TDS (R is 0.974, 0.99, 0.999, 0.982, respectively) (Table 1); the anions of Cl^-, SO_4^{2-} and B^- also exhibited aa significant positive correlation with TDS (r score). The correlation between NO_3^- and TDS was relatively weak (r = 0.766). Accordingly, Mg^{2+}, Na^+ and K^+ ions in groundwater showed a significant positive correlation with TDS (r = 0.986, 0.998, 0.959). Only Cl^- and SO_4^{2-} among the anions had a significant positive correlation with the TDS (r = 1, 0.989), while other anions exhibited no significant correlations.

Table 4. Correlation matrix of anions and cations in surface water and shallow groundwater of coastal region.

Index	pH	COD	TDS	Ca^{2+}	Mg^{2+}	Na^+	K^+	NH_4^+	Cl^-	F^-	SO_4^{2-}	Br^-	I^-	HCO_3^-
							Surface water							
COD	0.151	1												
TDS	-0.432	0.21	1											
Ca^{2+}	-0.455	0.119	0.974**	1										
Mg^{2+}	-0.458	0.103	0.99**	0.978**	1									
Na^+	-0.401	0.251	0.999**	0.97**	0.984**	1								
K^+	-0.425	0.293	0.982**	0.964**	0.954**	0.985**	1							
NH_4^+	0.002	-0.098	-0.067	-0.178	-0.018	-0.075	-0.218	1						
Cl^-	-0.432	0.197	1**	0.973**	0.992**	0.998**	0.979**	-0.055	1					
F^-	-0.540	-0.122	0.352	0.362	0.337	0.348	0.408	-0.163	0.348	1				
SO_4^{2-}	-0.51	0.181	0.988**	0.971**	0.974**	0.983**	0.984**	-0.136	0.987**	0.389	1			
Br^-	-0.361	0.233	0.906**	0.852**	0.869**	0.909**	0.899**	0.3	0.904**	0.525	0.905**	1		
I^-	-0.431	-0.31	0.255	0.146	0.311	0.23	0.109	0.775**	0.268	0.227	0.211	0.31	1	
HCO_3^-	0	-0.135	-0.584	-0.685*	-0.556	-0.594	-0.667*	0.581	-0.576	-0.296	-0.609	-0.561	0.488	1
NO_3^-	-0.239	0.336	0.766**	0.734*	0.742*	0.778**	0.754*	-0.001	0.763*	0.264	0.727*	0.738*	0.117	-0.37
							Groundwater							
COD	0.618**	1												
TDS	-0.409	-0.176	1											
Ca^{2+}	-0.785**	-0.458	0.166	1										
Mg^{2+}	-0.418	-0.185	0.986**	0.215	1									

Na^+	-0.369	-0.15	0.998**	0.113	0.979**	1								
K^+	0.279	-0.091	0.959**	-0.017	0.912**	0.969**	1							
NH_4^+	0.365	0.54	0.417	-0.119	0.458	0.458	0.641*	1						
Cl^-	-0.409	-0.176	1**	0.174	0.985**	0.998**	0.958**	0.408	1					
F^-	0.792**	0.454	-0.406	-0.682**	-0.379	-0.375	-0.317	0.412	-0.405	1				
SO_4^{2-}	-0.419	-0.172	0.989**	0.122	0.972**	0.991**	0.958**	0.37	0.986**	-0.414	1			
Br^-	0.212	0.445	0.293	-0.056	0.368	0.18	0.18	0.923**	0.289	0.003	0.277	1		
I^-	0.015	-0.047	0.116	0.069	0.223	0.105	-0.73	0.541	0.111	0.044	0.094	0.615**	1	
HCO_3^-	-0.259	-0.447	0.027	0.047	0.084	-0.375	-0.055	-0.164	0.014	-0.196	0.069	0.004	0.446	1
NO_3^-	0.212	0.445	0.293	-0.056	0.368	0.013	0.18	0.923**	0.289	0.003	0.277	0.004	0.615**	0.004

*Correlation significant at *a* = 0.05

**correlation is extremely significant at *a* = 0.01.

Analysis of water environment

Surface water quality

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The geochemical properties and the evaluation results of the surface water are illustrated in Table 2. The surface water quality was evaluated according to GB3838-2002. In the evaluation, three samples in the station (DBY01, DBY08, DBY09) were close to the sea water channel samples, and they were evaluated according to sea water quality standard (GB3097-1997).

The surface water quality was generally poor in all the assessed samples, belonging to IV or V quality grade (Table 2). F^- of DBY05 water exceeded the class V of GB3838-2002 surface water environment quality standard. F^- and COD of DBY08 sea water samples exceeded the class IV of GB3097-1997 seawater environment quality standard.

Shallow groundwater quality

The single factor evaluation method was adopted to evaluate the shallow groundwater, and groundwater quality standard (GB/T14848-2017) was

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taken for the reference (Figure 8). The shallow groundwater quality and data is shown in Table 4.

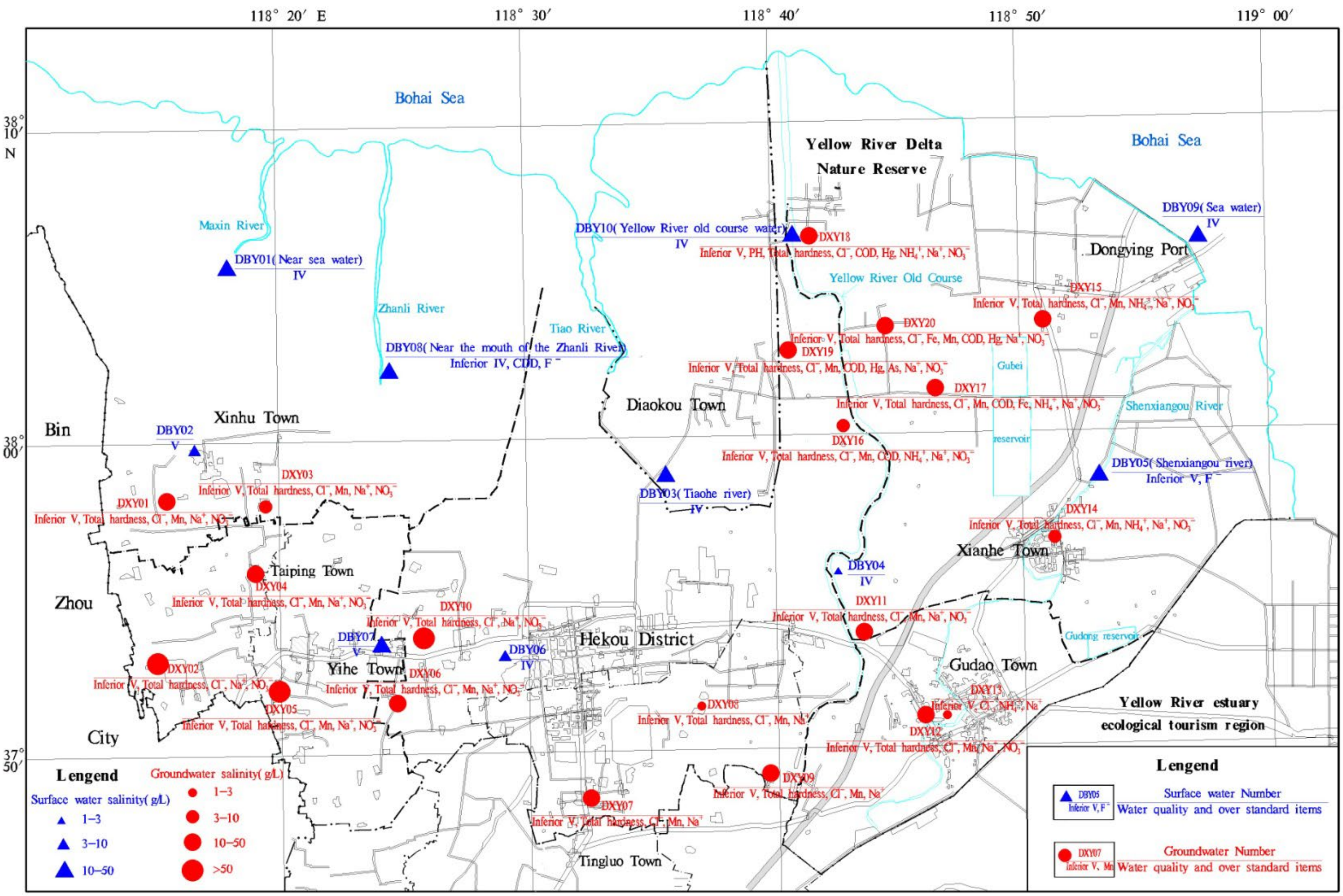

Figure 8. Spatial characteristics of salinity and pollution in surface water and shallow groundwater quality.

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All groundwater samples quality grades belong to Inferior V. The eight indicators exceeding the groundwater V quality standard included Cl^-, Na^+, total hardness, COD, NH_4^+, Hg, As and NO_3^-. The indicators exceeding the groundwater quality V standard included Cl^- and Na^+ in all samples, total hardness in all except the DXY13 sample, COD in samples of DXY16~DXY20, NH_4^+ in samples of DXY13~DXY18, Hg in samples of DXY18~DXY20, Arsenic in sample of DXY19, and NO_3^- in samples of DXY02~DXY06, DXY10~DXY12 and DXY14~DXY20.

Conclusions

The present study envisaged the dynamic monitoring and study of the hydrochemistry of the surface water and shallow groundwater in the northern part of the Yellow River Delta, China. The important conclusions of the study are as follows:

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1. The dynamic types of shallow groundwater in the northern part of the Yellow River Delta can be divided into two types: precipitation infiltration evaporation type and hydrological type. The precipitation evaporation type was mainly distributed in the coastal area, and the utilization degree of the Yellow River diversion was relatively low, which was the brine or salt water area. The groundwater runoff was slow, with precipitation infiltration and phreatic water evaporation as the main alternative mode. The hydrological type was distributed in the influence zone along the old Yellow River, and the groundwater level dynamic was related to the water level dynamic of the Yellow River together with the influence of meteorological and irrigation infiltration factors.

2. The surface water and shallow groundwater in the northern part of the Yellow River Delta exhibited a higher degree of mineralization, and the overall degree of mineralization was higher in the shallow groundwater. The shallow groundwater was of Cl-Na·K type, and it was a typical mixture or brine. The surface water changed to $Cl{\cdot}SO_4$-Na·K·Ca type, owing to the industrial and agricultural and city pollution. The hydro-chemical type was complicated. In surface water and shallow groundwater, cations of Mg^{2+}, Na^+ and K^+, and anions of Cl^- and SO_4^{2-} were found to be the main ions related to TDS.

3. In the northern part of the Yellow River Delta, the pressure of the water resources and water environment was relatively high, and the available water resources mainly came from the Yellow River. The overall water environment quality was low. Due to the oil production concentration area, large-area brine distribution area of Shengli Oilfield, petroleum enterprises, chemical and brine, urban sewage discharge and municipal solid waste gathering in this area, the deterioration of water environment quality was aggravated. For the sustainable development of the Yellow River Delta, it is necessary to strengthen the monitoring and management of the total discharge control of the oilfield, chemical industry and urban domestic water, and undertake effective measures to ensure that the industrial pollution sources meet the discharge standards. At the same time, it is also important to strengthen the land spatial planning and layout, reduce new projects of heavy and petrochemical industry, and increase the green and sustainable development industries.

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