Journal of Contaminant Hydrology xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Journal of Contaminant Hydrology

journal homepage: www.elsevier.com/locate/jconhyd

Using environmental isotopes along with major hydro-geochemical compositions to assess deep groundwater formation and evolution in eastern coastal China

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ARTICLE INFO

Keywords:
Hydrochemistry
Stable isotopes
Groundwater age
Groundwater development

ABSTRACT

Hydrochemical analysis and environmental isotopic tracing are successfully applied to study groundwater evolution processes. Located in eastern China, the Jiangsu Coastal Plain is characterized by an extensively exploited deep groundwater system, and groundwater salinization has become the primary water environmental problem. This paper provides a case study on the use of a hydrochemical and environmental isotopic approach to assess possible mixing and evolution processes at Yoco Port, Jiangsu Province, China. Hydrochemical and isotopic patterns of deep groundwater allow one to distinguish different origins in deep water systems. HCO_3^- is the dominant anion in the freshwater samples, whereas Na^+ and Cl^- are the dominant major ions in the saline samples. According to $\delta^{18}O$, $\delta^{2}H$ and ^{14}C dating, the fresh water is derived from precipitation under a colder climate during the Glacial Maximum (Dali Glacial), while the saline groundwater is influenced by glacial-interglacial cycles during the Holocene Hypsithermal. The $\delta^{18}O$, $\delta^{2}H$ and ^{3}H data confirm that deep groundwater in some boreholes is mixed with overlying saline water. The deep groundwater reservoir can be divided into a saline water sector and a fresh water sector, and each show distinct hydrochemical and isotopic compositions. The saline groundwater found in the deep aquifer cannot be associated with present seawater intrusion. Since the Last Glacial Maximum in the Late Pleistocene, the deep groundwater flow system has evolved to its current status with the decrease in ice cover and the rising of sea level. However, the hydraulic connection is strengthened by continuous overexploitation, and deep groundwater is mixed with shallow groundwater at some points.

1. Introduction

A coastal area is a vulnerable region for groundwater resources. Groundwater salinization is widespread throughout the world because of overexploitation (Amor et al., 2011; Monjerezi et al., 2011; Zhou et al., 2011; Giambastiani et al., 2013; Carreira et al., 2014). Natural hydrochemical evolution patterns have changed since the 1970s in the Jiangsu Coastal Plain in China due to the extraction of groundwater on a large scale (Huang and Lu, 2004; Zhou et al., 2011). Groundwater salinization has become the primary environmental water problem in this coastal area. Moreover, groundwater in the coastal plain faces a number of problems, including salinization due to mixing with present or ancient seawater, water-rock interaction, ion exchange, evaporation, etc. (Edmunds and Wright, 1979; Edmunds et al., 2003; Amor et al., 2011; Lin et al., 2011; Carreira et al., 2014). Classical hydrogeological studies are not useful for drawing conclusions regarding these problems. An adequate understanding of groundwater formation and evolution is essential for the sustainable exploitation of groundwater, but little attention has been paid to salinization processes in the coastal plain.

Recent studies have proven the usefulness of using a multi-tracer approach that combines both hydrochemical and isotopic data to identify groundwater origins and processes. Hydrochemical analyses are frequently applied to study the problem (Edmunds et al., 2003; Monjerezi et al., 2011; Carucci et al., 2012; Giambastiani et al., 2013; Zabala et al., 2015). Environmental isotopic tracers have been successfully applied in various evolutionary contexts. Stable isotopes ($\delta^{2}H$, $\delta^{18}O$), often combined with tracers of residence time (^{3}H, ^{14}C), are commonly used to investigate the hydrological conditions of recharge, runoff and discharge (Edmunds and Wright, 1979; Ha and Zhao, 1990; Negrel et al., 2011; Négrel et al., 2012), as evidence of the hydraulic connection among aquifers (Amor et al., 2011; Lin et al., 2011; Carucci et al., 2012; Carreira et al., 2014) and to discriminate the flow path (Bretzler et al., 2011; Bouragba et al., 2011; Hosono et al., 2011; Zhou

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https://doi.org/10.1016/j.jconhyd.2017.11.003
Received 15 June 2017; Received in revised form 1 November 2017; Accepted 4 November 2017
0169-7722/ © 2017 Published by Elsevier B.V.

et al., 2011; Siebert et al., 2012; Parker et al., 2012; Raidla et al., 2012).

In the coastal plain of Jiangsu Province, the aquifers containing fresh water formed during the early Pleistocene and Neocene, and have been subject to multiple sea-level fluctuations (Ha and Zhao, 1990; Li and Fan, 2009). Environmental isotopes (δ^2H, $\delta^{18}O$, ^3H and ^{14}C) were applied to trace the origins and processes involved to explain the groundwater problem (Ha and Zhao, 1990; Zhao and Liang, 1993; Xu et al., 2015), and hydrochemical analyses were used to decipher the origin and mechanism of groundwater salinization (Zhou et al., 2011; Xu and Liu, 2015). Previous research on groundwater in the coastal plain has mainly focused on shallow groundwater, while little attention has been paid to deep groundwater. Moreover, most deep groundwater samples were mixed with overlying water in the past, which complicated or distorted the isotopic and chemical compositions. Yoco is an important deep port under construction on the Jiangsu Coastal Plain. The excellent water quality of deep groundwater is an important basis for regional economic and social development. In this study, we propose to investigate the origin and evolutionary process of deep groundwater in the coastal aquifers in Jiangsu Province based on δ^2H, $\delta^{18}O$, ^3H and ^{14}C contents together with major chemical data. The specific aims are as follows: (1) assess the groundwater quality and flow pattern configuration in the deep aquifers, (2) identify a possible origin and evolutionary processes of deep groundwater, and (3) determine the primary source of deep groundwater salinization. This study may be beneficial to the long-term prevention of groundwater quality deterioration and enhance understanding of a groundwater salinization process in a complex and multi-layered aquifer.

2. Outline of the study area

2.1. Geological and hydrogeological setting

The Jiangsu Coastal Plain is in the East Jiangsu Province in China; Yoco Port is situated on the southern branch of the plain. The area consists of a thick series of alluvial sediments. The plain has a low slope ranging from 1 m to 5 m above sea level (a.s.l.). The plain area is approximately 32,500 km^2, with an approximate length of 450 km and a width of 150 km (Fig. 1). This study focuses on the Yoco Port area. The Rutai Canal River runs through the southern plain from west to east. The study area is in the northeastern part of the Yangtze block (Zhang et al., 2015), and the surface is covered by unconsolidated sediments from the Early Pleistocene and Holocene. The basement under the loose sediments is Mesozoic to Neogene strata. The Cenozoic deposit is approximately 500–600 m thick and is composed of materials derived from acidic silicate rocks. Quartz and feldspar are the dominant minerals. Calcite and iron manganese oxides are common. Illite, kaolinite and montmorillonite are observed in the plain.

The climate of the study area is generally humid subtropical, with a mean annual temperature of 14.8 °C and a mean annual rainfall of 1025 mm. The coastal river and lake water is generally salty because of seawater intrusion. Therefore, groundwater is exploited. Annual groundwater exploitation is approximately 1.1×10^7 m^3 (122 mm) in the study area. The uncontrolled extraction of groundwater in the Jiangsu Coastal Plain in China has led to a decrease in the groundwater level, flow field change, groundwater resource reduction and progressive groundwater salinization.

The coastal plain is characterized by several porous aquifer systems (Table 1; Fig. 1). The permeable layers consist of unconsolidated sands, which are separated by layers of poorly permeable silt clays. The stratigraphic sequence comprises 3 units (Ha and Zhao, 1990). The uppermost unit in the study area is a shallow porous water system, which contains a series of Holocene estuary or coast sediments. These sediments include fine sands and loamy clay that are laterally discontinuous, resulting in a highly heterogeneous aquifer system. The phreatic aquifer is recharged by local rainfall and discharges from the Rutai Canal River. The unconfined aquifer is generally 25 to 40 m deep, and the water table is at a depth of from 1 to 4 m. There is a middle porous water system underneath the phreatic aquifer, which formed during the Middle to Late Pleistocene. The middle system is composed of the confined aquifer I and confined aquifer II, and is composed of fine sands, silts and gravels. Aquifer I is generally at a depth of from 40 to 140 m, and the water table is from 2 to 3 m deep. Aquifer II is generally at a depth of from 220 to 270 m, and the water table is from 3 to 5 m deep. Both the shallow and middle systems are generally saline water. The lowermost unit is the deep porous water system, and consists of Neogene to Early Pleistocene fluvio-lacustrine sands, gravels, silts and clays. The deep water system includes confined aquifer III and confined aquifer IV and is the main reservoir of groundwater in the study area. Aquifer III is generally found at a depth of from 230 to 300 m, and its water table is from 10 to 50 m deep, with generally eastward deepening, while the aquifer IV is at a depth of from 350 to 450 m, and its water table is from 10 to 30 m deep. Our study focused on the deep water system. According to the water level monitoring data, the water table in aquifer III declined (Xu and Liu, 2015). The groundwater level depression cone was approximately 698 km^2, and Matang was the center of the cone, with a water table $>$ 40 m in depth (Fig. 1).

The hydraulic connection between the shallow groundwater and surface water is closely linked, and shallow groundwater shows strong regeneration, while deep groundwater flow is slow and shows weak regeneration.

3. Sampling and methods

The water samples were collected from boreholes, the river and the sea in October 2014 (Fig. 1) using standard sampling procedures (CGS, 2012). There were 33 sampling points, including 21 samples from the deep monitoring boreholes, 7 samples from the shallow boreholes, and 5 samples from the surface water (river and sea). All water samples were collected and stored in polyethylene plastic bottles. The 21 groundwater samples were used for major ion analysis, while all of the selected samples were studied using isotope analysis. The samples for major ion analysis were filtered, and those for cation analysis were acidified with HNO_3 (pH $<$ 2) before storage.

Samples for radiocarbon (^{14}C) analysis were obtained by precipitating $BaCO_3$ by the addition of excess $BaCl_2$ to 100–120 L of water previously brought to a pH $\geq$ 12 by the addition of NaOH. ^{14}C was determined radiometrically by liquid scintillation counting after conversion to benzene (Chen et al., 2003).

The major hydrochemical concentrations and isotopic compositions of all deep groundwater samples were measured (TDS, pH, Fe, Mn, Na^+, Ca^{2+}, Mg^{2+}, K^+, HCO_3^-, Cl^-, SO_4^{2-}, NO_3^-, δ^2H, $\delta^{18}O$, ^3H, ^{14}C) using standard methods (CGS, 2012). The pH, temperature and TDS were measured in the field using portable metres (HANNA, HI8424; THERMO scientific, ORION) and calibrated using a standard solution. The major hydrochemical concentrations (Na^+, Ca^{2+}, Mg^{2+}, K^+, HCO_3^-, Cl^-, SO_4^{2-}, NO_3^-, Fe, Mn) and stable-isotopes (δ^2H and $\delta^{18}O$) were determined at the Laboratory of the Nanjing Institute of Geology and Mineral Resources in China.

Major elements (Na^+, K^+, Ca^{2+}, Mg^{2+}, Fe, Mn, H_2SiO_3) were measured using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). Major anion concentrations (Cl^-, SO_4^{2-}, NO_3^-) were determined using Ion Chromatography. CO_3^{2-} and HCO_3^- were determined using acid titration. δ^2H and $\delta^{18}O$ were determined using EA-MAT253 isotope mass spectrometry. The δ^2H and $\delta^{18}O$ analyses adopted the international standards of the VSMOW (Vienna Standard Mean Ocean Water) (Monjerezi et al., 2011). The results for δ^2H and $\delta^{18}O$ had an error margin of ± 2‰ and ± 0.1‰, respectively.

The determination of ^{14}C and ^3H in the groundwater was carried out at the Karst Geology and Resources Laboratory of the Institute of Karst Geology, China. The ^{14}C ages were expressed as the year (a), and the tritium (^3H) concentrations were expressed in TU. The analytical precisions were ± 5% and 0.1 TU for the ^{14}C ages and ^3H, respectively.

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