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Groundwater hydrochemical characteristics and processes along flow paths in the North China Plain



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ABSTRACT

The North China Plain is one of the biggest plains in China, where municipal, agricultural and industrial water supplies are highly dependent on groundwater resources. It is crucial to investigate water chemistry and hydrogeochemical processes related to hydrogeologic settings for sustainable utilization of groundwater resources. Two hydrochemical profiles proximately along the groundwater flow paths were selected for hydrogeochemical study. Major components and ²H and ¹⁸O isotopes were analyzed in groundwater samples from the profiles. The study area was divided into three zones, including strong runoff-alluvial/pluvial fans in the piedmont area (Zone I), slow runoff-alluvial/lacustrine plain in the central area (Zone II), and discharge-alluvial/marine plain in the coastal area (Zone III). Major components of groundwater samples showed obvious zonation patterns from Zone I to Zone III. Total dissolved solid (TDS) concentrations gradually increased, and the hydrochemical type changed from HCO3-SO4-Ca-Mg and HCO₃-Cl-Ca-Mg types to HCO₃-SO₄-Na-Ca, SO₄-Cl-Na-Ca and SO₄-Cl-Na types from Zone I to Zone III. Abrupt increases in concentrations of Na⁺, Cl⁻ and SO₄⁻ in deep groundwater were observed around the depression cones, which indicated that overexploitation resulted in water quality deterioration. Calcite and dolomite precipitation occurred in Zone I of deep groundwater systems and shallow groundwater systems. Cation exchange was believed to take place along the entire flow paths. Gypsum tended to dissolve in groundwater systems. The depletion in D and ¹⁸O isotopes in deep groundwater was related to the recharge from precipitation in paleo-climate conditions in glacial or interglacial periods, indicating that renewal groundwater was very limited. Efficient strategies must be taken to preserve the valued water resources for sustainable development.

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1. Introduction

Groundwater is the major resources for drinking, irrigation and industry in arid–semiarid areas. It has been of great significance to investigate geochemical evolution of groundwater at basin scales due to its great helps in better understanding spatial and temporal distribution of groundwater chemistry and in efficiently managing groundwater resources for domestic, industrial and agricultural water supplies (Carrillo-Rivera et al., 2008; Krause et al., 2007; Ayotte et al., 2011; Hosono et al., 2009). Groundwater geochemical evolution is controlled by both natural processes and human impacts. In natural systems, specific hydrogeochemical processes occur in different hydrogeologic settings (Shen et al., 1993). In recharge areas, dissolution of minerals (including carbonates and

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silicates) dominates (Sung et al., 2012). Precipitation of secondary minerals prevails in discharge areas (Edmunds et al., 2006). On the other hand, seawater intrusion resulting from abstraction of groundwater in the coastal aquifer (Vandenbohede et al., 2009; Giambastiani et al., 2007; Ghosh Bobba, 2002; Sun et al., 2010), decline of water levels due to shallow groundwater pumping (Gao et al., 2007; Zhang, 2008; Yang et al., 2010), and nitrate introduction to shallow groundwater due to fertilizer usage (Hamilton and Helsel, 1995; Arnade, 1999; Cambardella et al., 1999; Krapac et al., 2002; Jalali, 2011) greatly changed geochemical processes and groundwater chemistry. In order to assess hydrogeochemical processes and geochemical evolution in the complex system at basin scale, many methods, including hydrogeochemical diagrams (such as Piper third-line diagrams), multivariate statistical analysis, water-rock interaction simulation and mineral phase equilibrium calculations have been intensively used (Barbecot et al., 2000; Beaucaire et al., 1995; Sikdar et al., 2001; Xue et al., 2000), as well as major components, stable isotopes, trace elements and redox indicators (Plummer et al., 1990; Rademacher et al., 2001;

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Edmunds et al., 2002; Condesso de Melo et al., 1999; Kretzschmar and Einsele, 1995; Adams et al., 2001).

As the largest alluvial plain in eastern Asia, the North China Plain (NCP) is one of the most water scarce areas in China, with about 450 m³ water resources per capita (Kreuzer et al., 2009). The water resource shortage has seriously impacted on economic development in the NCP (Jia and Liu, 2002; Wang et al., 2008). In the 1960s, many reservoirs were built in the front of the west mountain and drainage channels were built in the east plain, leading to the decline of water storage in aquifers and the dry-up of rivers. Due to depletion of the surface water, groundwater has been used as major water supply for agricultural, industrial and domestic needs since the 1970s (Zhang et al., 1992; Zhang et al., 2000). Hundreds of thousands wells were used to pump groundwater from both shallow aguifers and deep aguifers to a maximum depth of 600 m. In 2003, there was 2.95 billion m³ deep groundwater abstracted for agricultural, industrial, and municipal purposes (Shi et al., 2010). Excessive exploitation of groundwater has caused groundwater levels to fall at alarming rates, and led to numerous drawdown cones with drawdowns of up to 80 m in the centers of depression cones (Chen et al., 2005b). The decline in groundwater levels has greatly changed the natural groundwater flow system, including recharge, runoff and discharge conditions (Wang et al., 2008; Zhang et al., 1997, 2000; Fan, 1998; Xia et al., 2004). Therefore, many investigations have been taken to delineate groundwater flow conditions and evaluate sustainable usage of groundwater resources in the NCP (Zhang et al., 2009; Zhang and Fei, 2009; Zhang, 2005; Zhang, 2008; Wang et al., 2009a,b; Yang et al., 2010; Xia et al., 2004; Wang et al., 2008; Shi et al., 1998; Jia and liu, 2002; Fei et al., 2009; Fan, 1998; Chen et al., 2005b).

The decline in groundwater levels may have caused severe changes of hydrochemical characteristics and geochemical processes in aquifers. Seawater intrusion was observed into coastal aquifers due to excessive exploitation (Han et al., 2011; Sun et al., 2010; Xue et al., 2000), which greatly changed natural hydrogeochemical zones from the alluvial to the coastal plain (Liu. 1999: Zhang et al., 2000). In nine cities of the Hebei plain with a mass exploitation of groundwater, groundwater deterioration was found (Chen et al., 2005a). The deterioration was possibly due to either agricultural pollution (Hu et al., 2005) or mixing of saline water from underlying aquifers (Chen et al., 2003; Zhang, 2005) being dependent of locations. However, changes of groundwater chemistry and related geochemical processes along the flow path are not well evaluated, which would help in better developing suitable utilization strategies for groundwater resources in the plain impacted by intensive groundwater abstraction.

The main objectives of this study are to (1) investigate water chemistry and isotope characteristics in typical hydrogeochemical zones of the plain, (2) evaluate chemical evolution of groundwater along the flow paths, and (3) assess hydrogeochemical processes in different hydrogeochemical zones of the plain.

2. Regional hydrogeology

2.1. Geological settings

The North China Plain (NCP) is located in the eastern part of China with the longitude between $112^{\circ}30'$ and $119^{\circ}30'$ E, and the latitude between $34^{\circ}46'$ and $40^{\circ}25'$ N, a total area of approximately 13.90×10^4 km², and the population of about 107.8 million. Lying between the west of Bohai Bay and the east of the Taihang Mountains, the NCP is bound to the south of the Yanshan Mountains and to the north of the Yellow River (Fig. 1).

The NCP is a large Mesozoic and Cenozoic sedimentary basin with the Sinian bedrock as a basement, which is controlled by

the North China fault depression. The underlying geology includes the neritic deposits of Sinian, Cambrian, Ordovician and late Carboniferous, terrestrial-marine deposits of the Cenozoic and Permian, and continental deposits of Cenozoic.

As affected by the new tectonic movements (including volcanic eruptions and seismic activities), the Yanshan Mountains and the Taihang Mountains are gradually uplifting, while the NCP is relatively declining since the Tertiary. Transgressions have frequently occurred in the eastern coastal areas. Alluvial and fluvial sediments originating from middle and lower reaches of the Yellow River, the Haihe River, the Luanhe River and their tributaries formed sedimentary aquifers in the Cenozoic basin. The sediment thickness of the Cenozoic formation is up to 1000–3500 m, with the Quaternary deposits ranging between 200 and 600 m. The Quaternary sediments are dominated by fluvial deposits in the piedmont plain, alluvial and lacustrine deposits in the central plain, and alluvial deposits with interbedded marine deposits in the littoral plain (Chen et al., 2003), which constitute the major aquifers for water supply in the NCP.

2.2. Hydrogeological settings

The NCP accessible groundwater mainly occurred in the Quaternary sediment aquifers. The regional Quaternary aquifers consist of fluvial fans, alluvial fans and lacustrine deposits (Chen, 1999; Zhang et al., 2000). From the top to the bottom, sediments can be divided into four aquifer groups according to the lithologic properties, geological age, the distribution of aquifers and aquicludes, and hydrodynamic conditions (Chen et al., 2003). The depth of the first aquifer group (shallow unconfined aquifer) ranged between 10 and 50 m, with coarse-grained sand in the piedmont area to fine-grained sand in the littoral plain. The second aquifer group was a series of shallow semi-confined aquifers with the buried depths 120-210 m, with sandy gravel, medium to fine sand (Chen, 1999). The second group was the major aquifers for groundwater exploitation for agricultural irrigation. The third aquifer group, underlying the second aquifer group, had lower boundary between 170 and 350 m (Zhang et al., 2009; Zhang, 2005; Yin and Sun, 1995). This formation consists of sandy gravel in the piedmont area and medium to fine sand in the central and littoral plain. The forth aquifer group lay below 350 m with a thickness of 50-60 m, which consists of cemented sandy gravel and thin layers of weathered sand (Zhang et al., 2000). According to groundwater exploitation and aquifer distribution, groundwater can be divided into shallow groundwater and deep groundwater (Zhang et al., 2009). Shallow groundwater mainly occurred in the first aquifer group (shallow aquifers), while deep groundwater in the latter three groups (deep aquifers).

Accordingly to land morphology, sediment characteristics, and groundwater flow condition (Chen, 1999; Zhang et al., 2000; Chen et al., 2003; Zhang, 2005; Zhang and Fei, 2009), the study area can be divided into three hydrogeochemical zones, including strong runoff-alluvial/pluvial fans in the piedmont area (Zone I), slow runoff-alluvial/lacustrine plain in the central area (Zone II), and discharge-alluvial/marine plain in the coastal area (Zone III) (Fig. 2). The alluvial-pluvial area is distributed as a ribbon in shape along the Yanshan Mountains in the north and the Taihang Mountains in the west. The aguifers in Zone I are mainly composed of clayey gravel and medium-coarse sand, with a high permeability. Groundwater was mainly recharged by means of lateral flow from mountain area and vertical infiltration from rivers and irrigation return (Wang et al., 2009a,b). Groundwater flow velocity ranged between 0.013 and 0.26 m/d in Zone I, evaluated from the data by Zhang and Fei (2009). The central alluvial/lacustrine plain is formed by alluvial-lacustrine sediments, which are mainly composed of clay, silty clay, and fine-medium sand (Zhang, 2005). From the west to

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