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Groundwater salinization processes and reversibility of seawater intrusion in coastal carbonate aquifers

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SUMMARY

Seawater intrusion (SWI) has led to salinization of fresh groundwater reserves in coastal areas worldwide and has forced the closure of water supply wells. There is a paucity of well-documented studies that report on the reversal of SWI after the closure of a well field. This study presents data from the coastal carbonate aquifer in northeast China, where large-scale extraction has ceased since 2001 after salinization of the main well field. The physical flow and concomitant hydrogeochemical processes were investigated by analyzing water level and geochemical data, including major ion chemistry and stable water isotope data. Seasonal water table and salinity fluctuations, as well as changes of δ^2H–$\delta^{18}O$ values of groundwater between the wet and dry season, suggest local meteoric recharge with a pronounced seasonal regime. Historical monitoring testifies of the reversibility of SWI in the carbonate aquifer, as evidenced by a decrease of the Cl^- concentrations in groundwater following restrictions on groundwater abstraction. This is attributed to the rapid flushing in this system where flow occurs preferentially along karst conduits, fractures and fault zones. The partially positive correlation between $\delta^{18}O$ values and TDS concentrations of groundwater, as well as high NO_3^- concentrations (>39 mg/L), suggest that irrigation return flow is a significant recharge component. Therefore, the present-day elevated salinities are more likely due to agricultural activities rather than SWI. Nevertheless, seawater mixing with fresh groundwater cannot be ruled out in particular where formerly intruded seawater may still reside in immobile zones of the carbonate aquifer. The massive expansion of fish farming in seawater ponds in the coastal zone poses a new risk of salinization. Cation exchange, carbonate dissolution, and fertilizer application are the dominant processes further modifying the groundwater composition, which is investigated quantitatively using hydrogeochemical models.

1. Introduction

Increased groundwater exploitation to meet the ever-growing fresh water demands has resulted in seawater intrusion (SWI) in coastal aquifers around the world. Karst terrains cover roughly one-tenth of the earth's continents (Drew, 1999) and groundwater from karst aquifers contributes significantly to the water demand in many coastal regions. Due to their importance, coastal karst aquifers have been extensively studied in, for example, America (Fleury et al., 2007), Spain (Martinez-Santos et al., 2005), Morocco (Bouchaou et al., 2008; El Yaouti et al., 2009), Mexico (Escolero et al., 2007), Israel (Kafri et al., 2007), Greece (Panagopoulos, 2008), and Croatia (Biondić et al., 2006). Well-documented case studies published in the literature are mostly from limestone and dolomite aquifers of Mesozoic to Cenozoic age, e.g. along the Mediterranean coast (Magaritz et al., 1984; Fidelibus et al., 1993), the Caribbean coast (Plummer, 1977; Back et al., 1979; Scott et al., 2002), and the Atlantic coast (Renken et al., 2002). These rocks were subjected to intense karstification that reached several hundred meters deep (Cita and Ryan, 1978; Bicalho et al., 2012), and may have high secondary (i.e. formed post-deposition) permeability (White, 2003). The resulting permeability distribution is thereby different from Paleozoic limestones and dolomites, which typically have very low secondary permeability (White, 2003). Apart from a few Chinese studies (e.g., Fan, 1984; Wu et al., 1994), it would appear that no studies have focused on

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salt water intrusion in these older systems, of which the coastal karst systems in northeastern China form an example. They developed in carbonate rocks of Ordovician and Cambrian age (Li et al., 2006), and it may be expected that their low permeability causes SWI to occur preferentially along faults and fracture zones.

Moreover, the dual-porosity nature of such mediums may have an impact on the reversibility of SWI, as seawater may enter the stagnant zones of the aquifer during stages of SWI, which then becomes slowly released by diffusive mass transfer after the more mobile parts of the aquifer have been freshened. Studies that document the reversal of seawater intrusion are quite rare in the literature and mainly constrained to technical reports. SWI reversibility has been identified in the Biscayne Aquifer near a well field in Broward County in Florida, USA (Dunn, 2000). There a seaward movement of the freshwater interface was recorded during a period of declined abstraction between 1979 and 1994, and a lag time was observed between the rise of water levels in the aquifer and measured salinities. The lag time ranged from 1.5 to 4.5 years in the wells analyzed and it was found to increase with decreasing permeability and increasing distance from the coast (Dunn, 2000). A seasonal dependence of SWI has also been reported for limestone aquifers in the Mediterranean region and in England (Giordana and Montginoul, 2006). A review of selected cases by these authors showed that the aquifer permeability and recharge rates form the dominant controls on the timescale of SWI reversibility. Giordana and Montginoul (2006) further found that in some sedimentary aquifers salinization by SWI persisted, leading to exploitation of groundwater of a lower quality.

This study reports on the coastal carbonate aquifer of Dalian region, where groundwater salinization occurred as early as 1964. Most of the previous research efforts in the study area have focused on identifying the extent of salinization near the Daweijia well field, which formed one of the largest groundwater sources for Dalian City before 2001 (Fan, 1984; Wu et al., 1994; Zou et al., 2001, 2004). The increase in groundwater salinity has been attributed to SWI caused by over-abstraction (Lü et al., 1981). However, relatively subtle increases in groundwater salinity in coastal aquifers like those observed in the Dalian area are not always due to SWI, and it cannot be excluded that the increase of groundwater salinity was not only caused by SWI, but also by human activities (e.g. agricultural irrigation, fish farming) and droughts.

To determine groundwater salinization sources and processes, and assess SWI reversibility, salinity and water level data from previous studies have been synthesized, and were complemented by newer chemical and isotope data collected after closure of the well field in 2001. Major ion concentrations combined with isotope data have successfully been used for this purpose in other studies (e.g., Vengosh and Rosenthal, 1994; Edmunds, 1996; Vengosh et al., 1999; Daniele et al., 2013). These data can also be used to understand the water–rock interaction processes associated with mixing, such as mineral dissolution/precipitation and ion exchange (Wigley and Plummer, 1976; Back et al., 1979; Nadler, 1980; Hanshaw and Back, 1985; Sanford and Konikow, 1989; Fidelibus and Tulipano, 1991; Stuyfzand, 2008).

This study is the first to comprehensively characterize the groundwater flow system in the coastal carbonate aquifers in northeastern China. The Dalian area provides an interesting case study as salinization became so severe that the Daweijia well field was forced to shut down in 2001. The main objectives were to (i) develop a conceptual model of the groundwater system of the Dalian region as an exemplar of a coastal karst aquifer in carbonate rocks of low permeability, (ii) understand the hydrochemical and hydrodynamic conditions after closure of the Daweijia well field, and (iii) document the temporal changes of the extent of SWI.

2. Study area

Dalian is located in the southern tip of Liaodong Peninsula, and is one of the most important industrial regions in China. The study area is located between 121°37′ to 121°44′ east and 39°10′ to 39°14′ north, and has an area of approximately 66 km^2 (Fig. 1). It is centered around the Daweijia well field of Dalian City, in the Liaoning Province in northeastern China. Elevation ranges from 121 m above sea level (m.a.s.l.) in the north, 140 m.a.s.l. in the east, and 320 m.a.s.l. in the south, to 1–2 m.a.s.l. at the coast in the west.

The area is characterized by a temperate semi-humid monsoon climate. The mean annual air temperature is 10.5 °C, mean annual rainfall (averaged from 1956 to 2000) is 613 mm (Li, 2004) and mean annual potential evaporation is 1548 mm (Yang, 2011). As much as 60–70% of the annual precipitation falls between June and September. The Daweijia River has a length of 15.5 km, and originates in the eastern part of the study area (Fig. 1). It has an annual average discharge of approximately $14 \times 106\ m^3$/year (Li, 2004), and has a seasonal flow regime.

Geologically, the area is part of an opening basin facing the Bohai Sea (Wu and Jin, 1990). The strata outcropping in the Daweijia area include Ediacaran argillaceous limestone, shale, slate and metamorphic rocks, Cambrian–Ordovician limestone and dolomitic limestone, Cambrian sandstone and shale, and Holocene and Pleistocene sediments (Fig. 1). The composition of strata is given in Table 1. The Cenozoic sediments in the Daweijia area are mostly underlain by the Cambrian–Ordovician limestone in the south and east and Cambrian sandstone and shale in the north.

The tectonic structures of the study area are well developed with faults and folds in various directions. There are two groups of faults developed in this area, including a NE–SW group (F1 and F2 in Fig. 1) and an E–W group (F3 and F4 in Fig. 1). Generally, karst development becomes less with depth, transitioning from caves at shallower depth to fissures in deeper parts (Li et al., 2006). Although the karst is well-developed in the upper carbonate rocks (0 to −40 m.a.s.l.), the presence of clay-rich sediments in this depth range can restrict water circulation (Zhao, 1991). The main aquifers in the study area are composed of pure and thickly-bedded limestone and dolomitic limestone in the lower Ordovician and upper-middle Cambrian sections between −40 and −70 m.a.s.l (Lü et al., 1981; Zhao, 1991). Karst here is less-developed than at shallower depths, but the absence of clay results in better hydrodynamic conditions (Zhao, 1991). The most productive carbonate aquifers are located in the valley along Daweijia River.

A Quaternary aquifer of 10–40 m thick sits on top of the carbonate aquifers, and the two aquifers are hydraulically connected (Jin and Wu, 1990). The Quaternary aquifer is mainly composed of alluvium and marine sand and gravel layers. The alluvium deposits are mainly distributed along the valley of Daweijia River, and are composed of sandy loam with a thickness of 2–10 m in the upper part, and sandy gravel with a thickness of 5–20 m in the lower part (Lü et al., 1981). The main composition of these deposits is quartz sandstone and limestone. Near the coast, the carbonate aquifer becomes semi-confined, and separated from the overlying Quaternary aquifer, by an up to 10 m thick marine clay layer (Lü et al., 1981). The marine clay layer has a thickness varying from 2 m inland to 10 m at coast within 3.5 km away from the coast (Lü et al., 1981).

The Daweijia well field is located near the fault zone and provided water to Dalian City. Pumping occurred at 3 locations from a total of 5 wells (2 at CG2, 2 at CG3, 1 near QG9, Fig. 1) with depths (bottom of the screen) ranging between 90 and 128 m (Lü et al., 1981). The screened intervals of these wells are mainly from 70 to 120 m depth below ground surface. During the periods of intensive withdrawal (1977–1984) groundwater salinity rose and water