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# Hydrochemical changes due to intensive use of groundwater in the carbonate aquifers of Sierra de Estepa (Seville, Southern Spain)

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#### SUMMARY

The carbonate aquifers of Sierra de Estepa, situated in southern Spain, are undergoing intensive groundwater exploitation. Consequently, the volume of pumping surpasses the average recharge for periods of several consecutive years. Under such conditions, nearby springs have either dried up or only function during short time periods, after very rainy episodes followed by long droughts. During the brief periods when the springs are active, their water and the water extracted by pumping are calcium bicarbonates, with a spatial-temporal variability of their physico-chemical characteristics that is mainly conditioned by the degree of functional karstification of each system. When the springs are inactive, the pumping water gradually increases in salinity and becomes HCO<sub>3</sub>ClCaNa, ClHCO<sub>3</sub>NaCa and ClNa. Under the new conditions caused by so much pumping, the main factors determining the hydrochemical changes are the mixing of waters and the subsequent reactions of dissolution-precipitation between (1) the recharge coming from rainwater, (2) the hypersaline inputs from the clay-evaporite aquitards situated on the edges and at the base of the aquifer, and (3) the water stored in each aquifer. The hydrochemical information acquired allowed us to characterize and model the groundwater of these aquifers, to study the causes of its great spatial and temporal variability, and explain the influence of exploitation. This research shows that making sustainable use of water resources associated with carbonate aquifers calls for sound knowledge of the relationship between the aquifer and other bodies of groundwater or surface water, the hydrochemical quality of these possible inputs, and the vulnerability of the aquifer to exploitation, which in turn is conditioned by the ratio between water reserves and recharge.

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#### 1. Introduction

Before submergible electropumps came into use and today's techniques of well drilling were developed, groundwater was a practically non-exploited resource, and its use was sustainable. In the past five decades, however (Burke et al., 1999; Llamas and Custodio, 2003; Shah et al., 2007), the situation has changed drastically. At present, groundwater is one of the most exploited natural resources below the earth's surface (Zektser and Everett, 2004). Depletion of groundwater, often accompanied by a deterioration in water quality, is a common environmental problem in areas of North America, Northern Africa, the Arabian Peninsula, Northern China, India and Southeast Asia (Llamas and Custodio, 2003; Van der Gun, 2012; Wada et al., 2010). In Europe, the main problems

deriving from the intensive exploitation of aquifers are concentrated in the east and the south of Spain, where semi-arid climatic conditions prevail. In this region, unlike others, most aquifers plagued by problems due to intensive use are aquifers of a carbonate nature. In this sense, we may underline that around the Mediterranean Basin 50% of the population is supplied through groundwater associated with carbonate aquifers (Margat, 2008), as opposed to the 25% world average (Ford and Williams, 2007).

When an aquifer is exploited, pumping produces a decrease in the water reserves, a reduction in natural discharge and/or an increase in recharge from other bodies of water (Bredehoeft et al., 1982). Hence, it is important to stress that the management and hydrological planning of an aquifer will be conditioned, among other aspects, by its renewal period (Custodio, 2003; Margat et al., 2006; Margat and Thauvin, 1989; Martos-Rosillo et al., 2014), by the possible deterioration of the water quality as a consequence of its intensive exploitation (Vrba, 2003) and by environmental problems stemming from the new hydrodynamic situation (Sophocleous, 2003). Margat et al. (2006) define renewal period







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as the quotient between the volume of the aquifer storage (reserves) and the average recharge. For these authors, it is a comparative concept, not absolute, and it is conditioned by numerous factors such as aquifer thickness, drainable porosity and climatic parameters (especially rainfall). In this regard, various authors propose different groundwater management strategies depending on the renewal period for each aquifer (Margat and Thauvin, 1989; Margat et al., 2006; Martos-Rosillo et al., 2014).

Water resources associated with carbonate aquifers are strategic, supplying domestic water and serving as a resource for irrigation and industry (Ford and Williams, 2007). Moreover, karstic systems contribute significantly to increase the biodiversity presented in both, surface and underground (Goldscheider, 2012).

The particular hydrogeological characteristics of carbonate aquifers, with heterogeneous porosity and permeability (White, 1999; Worthington, 1999; Hartmann et al., 2014), are made manifest in the processes of infiltration (diffuse and/or concentrated), fluxes in the saturated and non-saturated zones (rock matrix and network of fractures and karstic conduits) and discharge (diffuse and/or concentrated) (Bakalowicz, 2005; Kiraly, 2003). Such conditioning factors can give rise to problems involving carbonate aquifers much more quickly than when the exploitation involves a detritic aquifer.

In carbonate aquifers with a well developed network of conduits, the zone of "dynamic storage" (the one above the level of drainage of the principal springs) is drained very quickly, above all if it is subjected to exploitation, given that the influence of pumping is compounded by the high capacity of these aquifers to empty via karstic springs. The long and frequent dry periods that characterize arid and semi-arid regions and the exploitation of these aquifers favor changes in the direction of the underground flow, even when they are exploited below average recharge. Under these circumstances, if the aquifers are in contact with the sea or with bodies of surface or groundwater of poor quality, contamination results. In turn, during periods when the springs remain inactive, serious environmental problems may arise, affecting the ecosystems associated with the natural dischange of these systems.

The hydrochemical processes in carbonate aquifers in a natural regime and their hydrogeological implications are well documented (Busenberg and Plummer, 1982; Cardenal et al., 1994; Moral et al., 2008; White, 1988), yet the same is not true of research about the deterioration of groundwater quality caused by its intensive use. In most cases the focus is on coastal aquifers undergoing advancement of the saline front (saltwater intrusion into coastal aquifers) (Calvache and Pulido-Bosch, 1994; Grassi and Tadolini, 1991; Pulido-Leboeuf, 2000; Sánchez-Martos et al., 1999). There are reports of contaminated water entry affecting agricultural activity (Edmunds and Walton, 1983; Llamas et al., 1992). There are also a few detailed papers about the deterioration of water quality due to the influx of poor-quality water (with high salinity) coming from aquifer or aquitard materials in contact with the carbonate aquifer exploited. In this sense, Rosenthal (1988) studied the increasing flow of Ca-chloride brines from deep-seated and confined reservoirs as a result of overexploitation of groundwater in Israel. This author determined the spatial distribution of areas where salinity increased as a result of the inflow of deep brines into the aquifers. Pulido-Bosch et al. (1995) related the progressive deterioration of water quality, with significant increases in chloride, sulfate and sodium, with the groundwater mining of the Crevillente aquifer (Alicante, southeastern Spain). They studied the temporal evolution of temperature, electrical conductivity of water, major ions and saturation indexes of calcite, dolomite and gypsum. Near to here, Pulido-Bosch et al. (1998) described an increasing salinity degree with depth in the carbonate aquifer of Cid. The base of this aquifer is constituted by rocks with halite and gypsum. Finally, Reynauld et al. (1999) studied the evolution of piezometric levels, volume of pumped water, rainfall, and groundwater chemical composition in Pinchinad Graben aquifer (Mouans Sartoux, France) along four years. The authors observed that the drop in the piezometric level caused the deterioration of water quality because of the inflow of saline water from the Triassic materials.

The use of multiple research techniques in this work has allowed the modeling of hydrochemical processes and mixing of waters caused by the exploitation as well as to establish the relationships between changes in water quality, the recharge rate and the storage capacity for each investigated aquifer.

The zone investigated here takes in the carbonate aquifers of the Sierra de Estepa (Sevilla province, southern Spain), considered as typical models of aquifers subjected to an intensive use of groundwater for urban and agricultural supply (Martos-Rosillo et al., 2013). The abundant hydrogeological information generated by means of different research projects carried out by the Geological Survey of Spain (IGME, 2006) and the diverse exploitation/recharge and recharge/reserve ratios of the systems studied provide a very adequate arena for the approach we use, putting forth measures oriented to manage and plan the sustainable use of groundwater.

The work carried out allowed us to characterize and model the hydrochemical evolution of the groundwater and study the causes behind its noteworthy variability over space and time. In the framework of this study, it is apparent that identifying and understanding hydrochemical processes is key to modify the hydrodynamic conditions of aquifers in order to achieve their sustainable use.

#### 2. Study area

Sierra de Estepa is located in southern Spain, 120 km east of the city of Sevilla (Fig. 1). This range is a small carbonate massif formed by several outcrops of Jurassic limestones and dolostones. Although the total exposed permeable surface is relatively small (34 km<sup>2</sup>), the carbonate aquifers of Sierra de Estepa supply water to some 36,000 people and around 600 ha of irrigation land (Martos-Rosillo et al., 2013).

In Sierra de Estepa average precipitation during the studied period (1976–2006) and its associated standard deviation are  $500 \pm 150$  mm/year. The months with the highest rainfall are November, December and January. Three drought events occurred during the analyzed period (1983–1986, 1990–1995 and 2004– 2006). The annually-averaged air temperature is between 15.9 and 17.6 °C, while the average annual potential evapotranspiration is between 1190 and 1300 mm/year (1976–2006).

Geologically, Sierra de Estepa lies in the External Zone of the Betic Cordillera, where the more competent Jurassic limestones commonly constituted isolated ranges, surrounded by the Antequera-Osuna Unit. Most of this unit is a mélange, and has a mixed tectonic, halokinetic and sedimentary origin. It largely involves Triassic clays, anhydrite, gypsum, halite, marls, and a minor amount of Mesozoic-Miocene limestone and siliciclastic rocks. It is a heterogeneous unit with a chaotic organization where hardy blocks are enclosed in a matrix composed by clays and evaporites (Flinch et al., 1996; Pedrera et al., 2012; Ruiz-Constán et al., 2012). The presence of hypersaline water associated to this geological unit is historically well-known because it has been used to obtain salt since the Roman times (Pérez, 2004). The distribution of salt bodies and their associated hypersaline waters is heterogeneous and irregular within the Antequera-Osuna unit. The geochemical signature of the water in the Antequera-Osuna aquitard is mostly sulfated because of the gypsum prevalence.

The carbonate aquifers of Sierra de Estepa are composed by 200–300 m of massive limestone and dolomitic breccias at the

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