



## Factors controlling groundwater salinization and hydrogeochemical processes in coastal aquifers from southern Spain



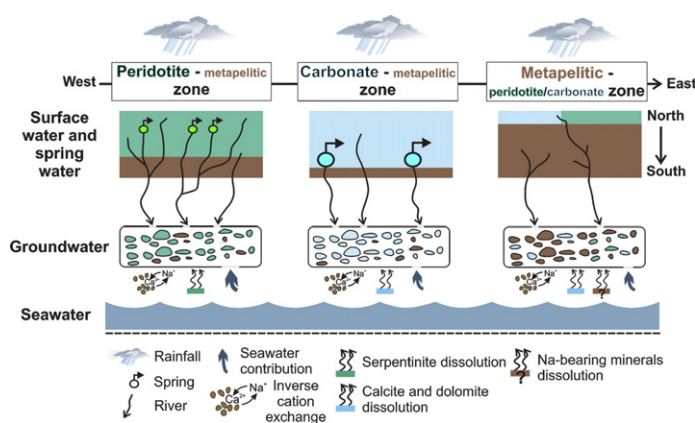
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### HIGHLIGHTS

- A full characterization of factors controlling GW mineralization and recharge sources has been performed.
- The proposed approach combines major and minor ions and water isotopes.
- Water-rock interaction and mixing processes govern GW geochemistry.
- Rock exposures greatly condition dissolved cationic fraction in GW.
- High surface water contributions to hydrochemistry of the W aquifer sector.

### GRAPHICAL ABSTRACT



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### ABSTRACT

In detrital coastal aquifers, seawater and surface water may interact with groundwater in multiple ways. Understanding the interference of water fluxes in this type of environment is essential to effectively manage the groundwater resources in water-stressed regions, such as the Mediterranean coastal fringe. In this research, the characterization of the main hydrogeochemical processes and the interaction between surface water and groundwater in the Marbella-Estepona coastal aquifers (southern Spain) have been carried out by means of the combined use of different hydrogeochemical indicators along with isotope data. The results show that the diversity of source lithologies (peridotite, carbonate and/or metapelitic) substantially conditions the groundwater geochemistry. The analysis of ionic deltas made it possible a preliminary screening of the geochemical reactions that occur in the Marbella-Estepona aquifers, while the Discriminant Analysis allowed for a consistent classification of sampled groundwater types. The dissolution of calcite and dolomite determines the chemical composition of the groundwater from the eastern sector that are more conditioned by the rainwater infiltration. The dissolution of magnesium-bearing minerals (predominantly forming peridotite rocks) is observed in groundwater samples from the western and central sectors, whose chemical composition showed a greater influence of surface water. The spatial analysis of  $rCl^-/Br^-$  in groundwater has permitted to corroborate that saline intrusion is negligible, hardly affecting to its original water quality. The irregularly distributed recharge by precipitation (seasonal effect) and the atmospheric circulation of cloud fronts (coastal/continental effect) explains why most of groundwater sampled is isotopically impoverished with respect to the rainfall signature. The isotope approach also suggests the hydraulic relationship between surface water and groundwater in the study site. A deeper knowledge of spatial hydrogeochemical variations in coastal groundwater and the influence of water sources

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over them are crucial for a sustainable groundwater management and global change adaptation in equivalent Mediterranean water-stressed regions.

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## 1. Introduction

Groundwater salinization is a common process affecting coastal aquifers worldwide (Barlow and Reichard, 2010; de Montety et al., 2008; Werner et al., 2013) as well as in several Mediterranean countries (Benavente et al., 2004; European Commission, 2004). Understanding the mechanisms involved in salinization process is a key factor to manage water resources in an efficient and sustainable way. Many researchers have dealt with the difficulties to determine the origin of salinization in coastal aquifers (Custodio, 1997; Bear et al., 1999; Araguás Araguás, 2003). Several sources have been identified, such as evaporite dissolution (Pulido-Leboeuf et al., 2003; Carol et al., 2009); downward leakage from surface saline water (Westbrook et al., 2005; Aunay et al., 2006) or wastewater (Ghabayen et al., 2006); deep brines or upward flow from deep saline water (Vengosh et al., 1999); and fossil (Tijani, 2004; Yamanaka and Kumagai, 2006;) or current seawater intrusion (Kim et al., 2003).

Saline intrusion is one of the main water quality issues in unconfined aquifers connected to the sea, particularly where excessive groundwater pumping may induce piezometric head decreases. In the case of confined or semi-confined aquifers, hydrogeological processes are more complex due to the existence of (i) clayey layers, which isolate, at least partially, groundwater from seawater, and (ii) a vadose zone where most of biogeochemical processes occurs (Groenewold et al., 1981). Therefore, saltwater intrusion may take place in both unconfined and confined aquifers (Groen et al., 2000; Bouchaou et al., 2009; El Yaouti et al., 2009).

Calcite dissolution is the main source of Alkalinity and  $\text{Ca}^{2+}$  in groundwater (Back et al., 1979). The dissolution/precipitation processes involving carbonate minerals substantially modify the water chemistry, but also its hydrogeochemical indices (Castillo and Morell, 1988). Detrital aquifers do not usually have a high content of carbonate minerals, such as calcite, aragonite or dolomite, although hydrolysis of calcareous matrix in sandy sediments may be responsible for increasing concentrations of  $\text{Ca}^{2+}$  and Alkalinity in groundwater (Jiráková et al., 2010). Besides the chemical reactions of several mineral species (calcite, dolomite, gypsum, etc.), the concentration of the main hydrochemical variables is very often controlled by cation exchange processes. These tend to compensate variations in the chemical composition of water, which significantly modifies the concentrations of major cations (Appelo and Postma, 1993; Martínez and Bocanegra, 2002; Andersen et al., 2005).

The geochemical evolution of groundwater depends on factors such as the residence time in the aquifer, the mineral species involved in water-rock interaction processes, and the occurrence of mixing processes with different sources of water (Appelo and Postma, 1993; Karanth, 1997).

The ionic ratios between different chemical species have frequently been used in groundwater salinization assessment in coastal areas (Vengosh and Rosenthal, 1994; Sánchez-Martos et al., 2002; Kim et al., 2003; El Moujabber et al., 2006). The relationships between major ions and  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  isotopes are widely used tools in studying complex mixing processes, involving rain, surface, seawater and groundwater, and therefore in determining the origin of salinity (Castillo and Morell, 1988; Capaccioni et al., 2005; Mollema et al., 2013; Mongelli et al., 2013). The present study proposes a methodological approach, based on the advances of hydrogeochemistry, for this purpose.

Costa del Sol area (southern Spain) has undergone intense demographic growth since the early 1960s, with the agricultural land cover

completely transformed into urban and tourist land uses. The drinking water supply depends mainly on surface water, which is stored in a water reservoir (La Concepción dam, in Fig. 1). Approximately 30% of water demand (12–14  $\text{hm}^3/\text{year}$ ; mainly for water supply of Marbella and Estepona municipalities) is satisfied by groundwater resources from aquifers; this percentage may, however, increase if several dry years occur consecutively, given the decreasing water levels in the reservoir. As a result of a drought between 1993 and 1995, groundwater overexploitation caused seawater intrusion processes (electrical conductivity reach  $\sim 45 \text{ mS/cm}$ ) that negatively impacted on groundwater quality (Pozos Reunidos, 1996).

In this coastal area, the dependence on groundwater resources is significant, particularly in low water availability periods (summer season) as it occurs in other Mediterranean regions (Iglesias et al., 2007). Therefore, there is a need to gain a deeper knowledge regarding water quality issues, specifically in a context of increasing groundwater use and decreasing natural recharge rate, which may give rise to seawater intrusion processes. In this situation, it is considered that groundwater governance plays a key role to involve all stakeholders towards a more integrated water resources management (Bressers and Kuks, 2004). Considering the latter assumptions, the present research aims to advance in i) the characterization of the hydrogeochemical processes taking place in coastal aquifers that are influenced by surface waters and seawater in multiple ways and ii) in the identification of water sources contributing to aquifer recharge. For these purposes, the Costa del Sol area has been selected as example of a highly urbanized and water-stressed Mediterranean coastal scenario.

## 2. Hydrogeological setting

The Marbella-Estepona aquifers are located in the west coast of Málaga province (southern Spain), from Estepona (west) to Elviria-Cabopino dunes (east) coastal locations (Fig. 1). It comprises a set of permeable outcrops of about 71  $\text{km}^2$ , although this area has been significantly reduced as consequence of the intense urbanization (Fig. 1). Shoreline morphology is relatively smooth, reaching maximum altitudes of 200 m a.s.l. (meters above sea level), nonetheless, more abrupt and higher reliefs limit the northern part of the studied area (Fig. 1). In this mountain range, constituted (from west to east) of Sierra Bermeja, Sierra Blanca and Sierra Alpujata reliefs, the catchment areas of the main rivers and intermittent streams (i.e. rivers Padrón, Guadalmanza, Guadalmina, Guadaiza, Verde, and Real) are found (Fig. 1). The interactions between surface waters and groundwater present a great seasonal and spatial variability (Argamasilla and Andreo, 2014), with a losing streamflow conditions in most cases. The studied rivers are protected by European Habitats Directive and have been recognized as Sites of Community Importance (MAGRAMA, 2006).

The average annual rainfall is 842 mm, although a precipitation gradient is observed (W to E), with values ranging from 950 to 723 mm in the urban settlements of Estepona and Marbella, respectively. The mean annual temperature is 18.8 °C, with average values of 12.9 °C in January and 25.6 °C in August.

The main aquifers are made up by two geological domains: the lower one (Pliocene) and the upper one (Quaternary) that overlie a basement of peridotite rocks, gneisses and marbles of the Alpujárride Complex of the Betic orogen and schists and graywackes of the Maláguide Complex (Fig. 1A), as well as marls and sandstones from Miocene formations (Aguado et al., 1990). Pliocene domain is composed of basal conglomerates and sands and sandy marls embedded in a

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