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## Geochemical processes controlling water salinization in an irrigated basin in Spain: Identification of natural and anthropogenic influence



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#### HIGHLIGHTS

- · Salinization in Lerma Basin was controlled by the dissolution of soluble salts.
- · Water salinization and nitrate pollution were found to be independent processes.
- High NO<sub>3</sub>, fresh groundwater evolved to lower NO<sub>3</sub>, higher salinity surface water.
- Inverse and direct geochemical modeling confirmed the hypotheses.
- · Salinization was a natural ongoing process slightly enhanced by land use.

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#### ABSTRACT

Salinization of water bodies represents a significant risk in water systems. The salinization of waters in a small irrigated hydrological basin is studied herein through an integrated hydrogeochemical study including multivariate statistical analyses and geochemical modeling. The study zone has two well differentiated geologic materials: (i) Quaternary sediments of low salinity and high permeability and (ii) Tertiary sediments of high salinity and very low permeability. In this work, soil samples were collected and leaching experiments conducted on them in the laboratory. In addition, water samples were collected from precipitation, irrigation, groundwater, spring and surface waters. The waters show an increase in salinity from precipitation and irrigation water to ground-and, finally, surface water. The enrichment in salinity is related to the dissolution of soluble mineral present mainly in the Tertiary materials. Cation exchange, precipitation of calcite and, probably, incongruent dissolution of dolomite, have been inferred from the hydrochemical data set. Multivariate statistical analysis provided information about the structure of the data, differentiating the group of surface waters from the groundwaters and the salinization from the nitrate pollution processes. The available information was included in geochemical models in which hypothesis of consistency and thermodynamic feasibility were checked. The assessment of the collected information pointed to a natural control on salinization processes in the Lerma Basin with minimal influence of anthropogenic factors.

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#### 1. Introduction

Salinization of water bodies represents a significant risk in water systems regarding suitability for irrigation (Tanji, 1990), other human uses (Peck and Hatton, 2003) or ecosystem health (Nielsen et al., 2003). There are many processes which can cause water salinization. For instance, the main sources of groundwater salinization in the US are natural saline groundwater, sea-water intrusion, halite dissolution, oil- and gas-field activities, saline seep, road salting and agricultural techniques (Richter and Kreitler, 1991).

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Groundwater salinization by agricultural practices has been widely

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reported all around the world (e.g., Koh et al., 2007; Stigter et al., 2006; Yuce et al., 2006). Specifically, irrigated agriculture represents an enhanced pressure on the hydrological system receiving irrigation return flows, with in general higher salinization rates under irrigated areas in comparison with rainfed areas (Johansson et al., 2009). Some examples of the reported processes that increase water salinity include the recirculation of irrigation water (Stigter et al., 2006), the application of agrochemicals (Kume et al., 2007), or the enhanced weathering of existing materials in the area (Kim et al., 2005; Koh et al., 2007). In some cases, natural salinization processes of water bodies are enhanced by irrigated agriculture, either by the transport of solutes in the water (Johansson et al., 2009) or by the depletion of aquifers and consequent concentration of salts (Chaudhuri et al., 2014). Irrigated agriculture may

induce salinization not only in groundwater, but also in surface waters (e.g., Duncan et al., 2008; Isidoro et al., 2006). In any case, the interaction between ground and surface waters adds complexity to the behavior of these hydrological systems. However, salinization in irrigated areas may be related to natural ongoing processes which may be, to a certain degree, enhanced by the addition of irrigation water (Carreira et al., 2014; Chaudhuri and Ale, 2014; Isidoro et al., 2006; Tedeschi et al., 2001) or a direct consequence of the irrigation itself (Stigter et al., 2006), since the dominating salinization processes are very sitespecific (Duncan et al., 2008). For instance, irrigation water enhances salinization processes in areas with available salts in the geological substrate, which are mobilized by the extra available water. In other cases, the continuous use of groundwater for irrigation may cause salinization due to the evapotranspiration that the groundwater body is suffering in successive pumping–irrigation–recharge cycles.

Thus, a deep knowledge of the local hydrogeological conditions and processes controlling salinization is required to understand the feasible actions to take in order to mitigate impacts on water resource systems. In line with this idea, two of the most useful tools to increase the knowledge of complex systems with several variables are multivariate statistical analyses (Hair et al., 1999) and geochemical modeling (e.g., Edmunds, 2009; Zhu and Anderson, 2002). Applications reported in the literature include the use of multivariate analysis to understand the main factor explaining the chemical evolution of waters in regional alluvial aquifers (Acero et al., 2013; Lorite-Herrera et al., 2008). For geochemical modeling, some examples of application are the use of inverse modeling approaches to elucidate the net geochemical reactions occurring in a water flow line from irrigation water to drainage water (Causapé et al., 2004a; García-Garizábal et al., 2014) or the use of direct modeling approaches to reproduce observed patterns in groundwater and to develop simulations in which different hypotheses can be tested (Kim et al., 2005; Stigter et al., 2006). However, both approaches are seldom applied to the same study area, which could improve the knowledge on the processes controlling the hydrological system.

In this context, the objective of the present study is to identify the main geochemical processes related to the salinization of waters in a headwater agricultural basin through the use of both multivariate statistical analysis and geochemical modeling. Specifically, the main aim is to elucidate if the processes controlling salinization of Lerma Basin waters (Merchán et al., 2013, 2014) are related to natural or to anthropogenic factors.

#### 2. Study site description

The Lerma Basin is a small hydrological basin (7.38 km<sup>2</sup>, Fig. 1), with 48% of its surface area under irrigation. It is located inside the municipality of Ejea de los Caballeros (Zaragoza, Spain) and is representative, regarding geology, hydrology and agronomy, of a wide range of land– water connected environments in the region (Causapé et al., 2004b).

According to the Spanish National Agency of Meteorology (AEMET, 2014), the climate in the area is Continental to Mediterranean, characterized by extreme temperatures and irregular and scarce precipitations. Temperatures may vary from below zero during winter to as high as 40 °C during summer, with an average temperature of 14 °C. Average annual precipitation during the hydrological years 2004–2011 was around 402 mm and concentrated in spring and autumn (Merchán et al., 2013). Summer and winter are generally dry, only wetted by occasional storms.

The geology in the Lerma Basin is represented by Tertiary and Quaternary materials (Fig. 1). The Tertiary materials appear as a bottom layer (66% of its surface) several hundreds of meters thick, composed of alternating gypsum, clay and silt materials of brown and gray colors, with occasional intercalations of fine limestone layers associated with gypsum (Causapé et al, 2004a; ITGE, 1988). This unit is locally covered by a Quaternary glacis (34% of the surface) up to 10 m thick, dominated by detrital sediments composed of gravels with some Tertiary limestone clasts, alternating with sands, silts and clays (ITGE, 1988). Tertiary materials are exposed due to the erosive effect of a network of gullies developed over the glacis.

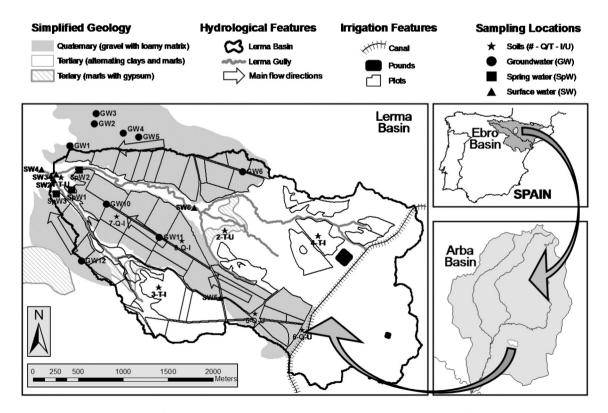


Fig. 1. Location, simplified geology, main hydrological and irrigation features, and sampling locations of the Lerma Basin.

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