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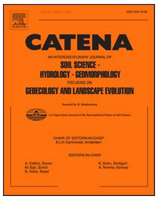

Studying hydrogeochemical processes to understand hydrodiversity and the related natural and cultural heritage. The case of Los Hoyos area (South Spain)

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ABSTRACT

The protection of geodiversity has been gaining interest during the last decades. However, the study of hydrodiversity has been much less developed than other aspects of the geological heritage (lithology, mineralogy, geomorphology, palaeontology). The objective of this work is to help with an inclusive definition of hydrodiversity by assessing its importance as part of geodiversity and how it can condition the natural and cultural heritage of a region. To that end, we have selected as a study area an evaporitic karst enclave of great geomorphological and environmental value located in southern Spain, named Los Hoyos, where diverse water features are found. Based on the hydrochemical and isotopic analysis of the water points, five main processes explain the hydrodiversity of the area: (1) the availability of minerals in the environment, (2) the residence time of groundwater, (3) the evaporation of water in the wetlands, (4) the common-ion effect (5) and the high ionic strength of groundwater. All these processes, directly related to the geology and geomorphology of the area (geodiversity), have given rise to diverse ecosystems (including protected wetlands), which enhance local biodiversity and geological forms (travertines), and are connected to the area's cultural heritage (salt extraction from the Paleolithic). This hydrodiversity is partially or totally responsible for a series of services to society related to its intrinsic, cultural, aesthetic, economic, functional, and scientific values. The case here presented exemplifies the importance of hydrodiversity in the natural and cultural heritage and highlights the need of advancing on the definition, promotion and protection of the hydrological heritage.

1. Introduction

The recognition of the need to protect geological heritage grew in the last decades of the 20th century. Thus, in the early '90s, several conferences on geoconservation took place, and the scientific community started to use the word "geodiversity" (Araujo and Pereira, 2018). Consequently, several definitions explaining the term came out (Duff, 1994; Osborne et al., 1998; Sharples, 1995, 1993; among others). Contrary to biodiversity, the definition of geodiversity is not unanimous (Serrano and Ruiz-Flaño, 2007a) since it considers different elements depending on the author. Most of the first descriptions of geodiversity did not include hydrological aspects (see Brocx and Semeniuk, 2007, and Gray, 2013 and reference therein), but with time other authors highlighted the value of water, in different forms, as an element of natural diversity (Brocx, 2008; Kozłowski, 2004; Serrano and Ruiz-Flaño, 2007b). Nowadays, one of the most widely accepted descriptions is that stated by Gray (2013): "*Geodiversity: the natural range (diversity) of geological (rocks, minerals, fossils), geomorphological (landforms, topography, physical processes), soil and hydrological features. It includes their assemblages, structures, systems and contribution to landscapes*". This definition updates the one previously made by the same author (Gray, 2004), including hydrology.

During the last decades, many countries have notably advanced in protecting their geological heritage, particularly since the development of the UNESCO geoparks initiative (Eder and Patzak, 2004; Jones, 2008). Lithological, mineral, geomorphological or paleontological diversity usually justifies the protection of these natural areas. On the contrary, hydrology and hydrogeology are often secondary elements in geological protected areas, although they can bring as much attention as the other mentioned aspects (Ruban, 2019).

The influence of geodiversity on biodiversity is undeniable as the geophysical elements are the drivers of the distribution of biota. Thus,

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authorities should incorporate abiotic diversity in biological conservation policies (Comer et al., 2015) and also in more interdisciplinary approaches focused on sustainable development (Brilha et al., 2018). One of the clearest examples of the biodiversity-geodiversity relationship is that geomorphological and hydrological aspects are the basis for many ecological classifications of wetlands and aquatic systems (Mitsch and Gosselink, 2015; Semeniuk and Semeniuk, 2016). From a broader point of view, any form of life is dependent on water, and different types of hydrological manifestations can host or promote unique species of flora, fauna, or microbiota. Thus, hydrological features of geodiversity are closely related to habitat provision. Still, they can also be connected to the other benefits that geology provides to society, including provisioning, regulating, supporting, and cultural services (Gray et al., 2013).

In part, for all of the above, in the last few years, the term “hydrodiversity” has appeared in the scientific literature, although with different meanings. The United Nations Development Program (PNUD, 2013) used the term to refer to the variety of water sources that are available for all humans, but most authors use it to describe the diversity of water elements in nature. While Graf (2001) referred to the aspects of naturalness for the hydrology of river channels, influencing habitats and society, other authors (Andreo et al., 2016; Ferrarin et al., 2014; Rosa et al., 2018) used the term hydrodiversity predominantly to explain the variety of wetlands according to their hydromorphological and hydrochemical characteristics. Most recently, some authors have considered both surficial water and groundwater as an element of hydrodiversity in assessing geodiversity at a regional scale (Araujo and Pereira, 2018; Bétard et al., 2018; Bétard and Peulvast, 2019; Fernández et al., 2020). Although these regional analyses value groundwater as an element of abiotic diversity, they only consider the existence of aquifers, but not their typology, the water quality, the hydrogeochemical processes, or the type of features directly related to them (springs, wetlands, rivers, travertine platforms, etc.). Even the Surf & Nature Alliance (2012) uses the term hydrodiversity in their “Manifesto for the protection of the waves” to talk about all the varied elements and places of the aquatic environment and, particularly, all surfable waves.

The use of the term hydrodiversity is therefore extensive and can potentially include a large number of features. However, few definitions are found in the literature, and they are primarily focused on the types of water elements. González-Trueba (2006) understand it as the variety of forms existing in the “hydrosphere” and the diversity of features and places made up of bodies of water in their different states. Meanwhile, Andreo (2010) broaden the meaning, defining hydrodiversity not only as the various manifestations of water but also as the diversity of its physicochemical properties. However, the variety of water elements goes beyond their typology or physicochemical characteristics. For example, the hydrodynamic responses of springs, rivers, wetlands, or even groundwater are unique and affect biodiversity and services to society. Tidal and wave characteristics in coastal areas are also elements of water diversity worthy of protection, as in the cases of the waves of Chicama, Peru, and Mundaka, Spain. Even changes in the physical state of water (freezing and melting) have ecosystem implications in cold climates (Vincent et al., 2008). In other words, water bodies are affected by hydro-bio-geo-physicochemical processes that provide hydrodiversity and play an essential role in the functioning of ecosystems, in addition to providing different types of benefits to humankind (Gil-Márquez et al., 2022).

This work aims to help define hydrodiversity by deepening into the origin of the diversity of water forms, particularly those related to groundwater. The objective is to identify all ecosystem and societal services provided by water features, understand the hydrogeochemical processes behind them, and propose an inclusive definition for hydrodiversity. To that end, we present the case of Los Hoyos area, an evaporite karst outcrop in South Spain of high geomorphological and landscape value, with very diverse water features. We analyse the variety of water elements (springs, wetlands, leakages) found in the area, the variability of mineralization (from fresh to brine waters), the different hydrogeochemical processes involved, and their ecological and social services. Together, all these aspects exemplify the importance of hydrodiversity and the need to consider it in conservation strategies.

2. Geographical and hydrogeological settings

“Los Hoyos” evaporitic karst area (Fig. 1) is part of an elongated outcrop of ENE-WSW direction in the northern part of Malaga province named “Trias de Antequera” (Peyre, 1974). It belongs to the Chaotic Subbetic Complex (CSC), a vast melange with a matrix formed by Upper Triassic (Keuper) clays and evaporite rocks containing blocks of diverse lithologies and ages (Vera and Martín-Algarra, 2004). Los Hoyos extends 20 km^2, between 700 and 900 m asl (above sea level), at the eastern Trias de Antequera. Its subcircular shape and other geomorphological observations suggest an origin related to halokinetic movements (Calaforra and Pulido-Bosch, 1999). The predominant lithologies in the area are multi-coloured clays and sandstones, evaporitic materials (gypsum and halite), carbonates (limestones, dolostones, dolomitic breccias), and subvolcanic rocks. The central part of the diapiric structure is occupied by gypsum, either as massive rocks or as polygenic breccia (Calaforra and Pulido-Bosch, 1993), covered and surrounded towards the edges by an olistrostromic megabreccia with a clayey-evaporitic matrix. Because of its high solubility, halite is not present at the surface, but its existence at depth is known (Carrasco, 1986). Around Los Hoyos area, there is terrain covered by Jurassic carbonates, Cretaceous marls and Tertiary marls, clays, and sandstones (Flysch). The CSC rocks are deformed so that their original stratigraphic relations are seldom preserved. Furthermore, the ground uplifting caused by the halokinetic processes resulted in a higher elevation of Los Hoyos (~50 m) regarding the surrounding Plio-Quaternary materials (placed between 700 -E border- and 750 m asl -W border-).

The exokarst development of Los Hoyos is remarkable, with numerous landforms, particularly karst depressions of different types (collapses, dissolution dolines, uvalas) and also sinkholes and swallow holes (Calaforra and Pulido-Bosch, 1999). Closed depressions placed at the central part of the study area, above 800 m asl, usually are small and present short-period floodings, as their bottom is above the groundwater table. The most remarkable example of that is the Vizcaino wetland (H3 in Fig. 1), located at 815 m asl. Dolines located towards the edges of the diapiric structure are more extensive and have longer hydroperiods. Among them, Grande and Chica lakes stand out, two wetlands protected as Natural Reserves by the regional government and also included in the Ramsar’s list. Grande lake (H1) is a permanent wetland with a maximum flooding surface of 8.57 ha and a maximum stage of 14.3 m. It occupies the bottom of a closed depression of 21.1 ha, whose bed locates at 780 m asl (Gil-Márquez, 2018). Chica lake (H2) is emplaced in an uvala (two coalescence dolines) with a maximum depth at 786 m asl. Its hydroperiod is almost permanent as it only dries in during extreme droughts, such as the one that occurred in the early ’90s (Rodríguez-Rodríguez et al., 2001). The maximum flooding extension is 8.03 ha, and the higher registered stage is 7.8 m. Its basin occupies 35.96 ha (Gil-Márquez, 2018).

According to the water table sketch of the area (CMA, 2005), illustrated in Fig. 1, groundwater predominantly flows from the central part of the area toward the western borders of the diapiric structure. One of the main discharge points of Los Hoyos area is the Aguileras spring (M1, at 787 m asl), located 365 m southeast of the centre of Grande lake. It is a permanent outlet with gentle flow rate variations and a mean value of 15 l/s (Rodríguez-Rodríguez et al., 2006). The spring emerges through a small gallery built to use groundwater to drive an old mill nearby. Despite the proximity of the spring to the lake and its location below the wetland water level, isotope determinations proved that the outlet drainage does not come directly from the lake (Gil-Márquez et al., 2016; Rodríguez-Rodríguez et al., 2007). Finally, a small seasonal outlet (Tunel leakage -R2-) connects to an ancient drainage tunnel west of Chica lake.

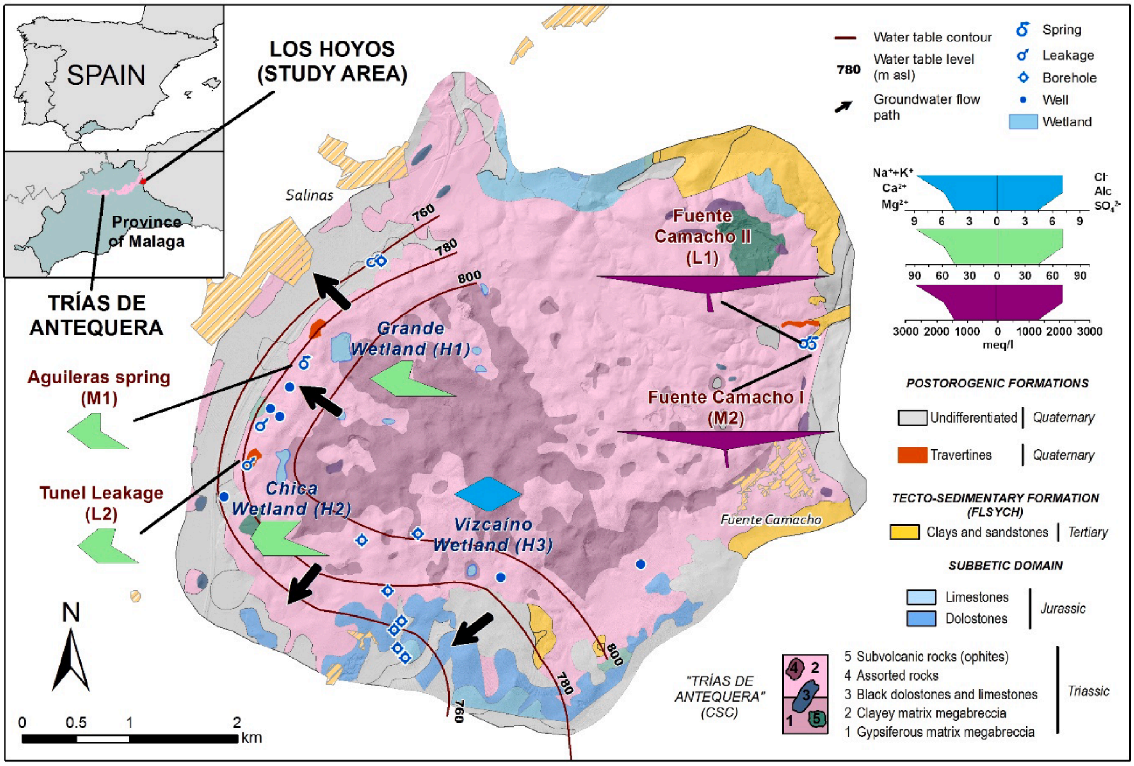

Fig. 1. Geological-hydrogeological map of Los Hoyos area. Modified from After Pineda-Velasco (1990). Water table sketch from CMA (2005).

On the eastern edge of Los Hoyos, near Fuente Camacho village (Fig. 1), discharge occurs through several brine outlets at a lower height (725–748 m asl). Two of those points were sampled during this study: Fuente Camacho I (M2), a spring located at 725 m asl, whose water has been used since ancient times for salt extraction, and Fuente Camacho II (R1), a small leakage placed 23 m above the first (748 m asl).

The climate in this area is temperate Mediterranean, characterized by a marked seasonal pattern of rainfall distribution, with a practical absence of precipitation during the summer. The mean annual rainfall for the period 1963/64–2015/16 was 609 mm (Gil-Márquez, 2018). The yearly mean temperature between 1975/76 and 2016/17 was 16.5 °C, with relatively warm summers (26.6 °C in July) and mild winters (7.9 °C in January). The mean annual evaporation between 2000/01 and 2016/17 was 1506 mm, most of it taking place between April and September, with a monthly maximum of 246 mm in July.

3. Methods

3.1. Analytical methods

3.1.1. Sampling routine and analytical methods

From September 2013 to September 2017, water level variations were registered in Grande lake and the Aguileras spring (OdysseyTM Capacitance Water Level). Additionally, single flow measurements were made in the outlet, so the water level record was transformed into flow-rate data series. A staff gauge was used for single lectures of the wetland stage. Samples for hydrochemical analysis were collected biweekly from the Aguileras, Fuente Camacho I and Fuente Camacho II outlets and from the Grande and Chica wetlands. Rain samples were also collected after precipitation events, and single samples were taken from the Vizcaino wetland and the Tunel leakage. Simultaneously with water sampling, *in situ* measurements of physic-chemical parameters were carried out, including electrical conductivity -EC- and water temperature (WTW® conductivimeter-thermometer 3310), and pH and dissolved oxygen -DO- (Hach® HQ40d). Data accuracy was ± 10% for flow rate, ±1% for EC, ±0.1 °C for temperature, ±0.1 mg/l of DO, and ± 0.1 units for pH.

Water samples were analyzed in the Laboratory of the Centre of Hydrogeology of the University of Malaga (CEHIUMA). Alkalinity was determined by volumetric titration with 0.02 N H_2SO_4 to pH 8.3 for CO_3^{2-} (if higher pH) and then to 4.45 for HCO_3^-. Major ion (Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, SO_4^{2-}, NO_3^-) hydrochemistry determination was performed by ionic chromatography (METROHM Compact 930 IC flex for cations and Compact 881 IC Pro for anions) with an accuracy of ± 2%. Samples with more than 1 mS/cm were diluted and filtered before being introduced into the chromatograph (inline and precolumn filters). Chemical data accuracy was checked by means of the charge balance error (CBE), obtained as the difference between cation and anion summations (Σcations – Σanions) divided by the summation of all the ions (Σcations + Σanions). Any samples with CBE higher than 5% were discarded, or up to 10% for brine waters (i.e., M2 and R1). The mean CBE for the whole sample set was 2.4%.

Water isotopes ($\delta^{18}O$ and δ^2H) of samples with EC values lower than 20 mS/cm were determined using a cavity ring-down laser spectrometer (Picarro® CRDS L2120-i) supported by an automatic sampler. Those samples with higher EC were analyzed using Isotope-ratio mass spectrometry (IRMS). In any case, isotopic data were referred to the Vienna-Standard Mean Ocean Water (VSMOW).

3.1.2. Isotope data treatment and modelling

The isotope activity ratio of water in brine solutions differs from the isotope concentration ratio because of the fractionation between free water molecules and those involved in the hydration sphere of the cations in water solution (Horita, 1989; Taube, 1954). The conversion of $\delta^{18}O$ and δ^2H data from activity scale to concentration scale was done using the "activity correction" defined by Sofer & Gat (1972, 1975):

$$\Delta\delta^2H = mNaCl\cdot(-0.4) + mMgCl_2\cdot(-5.1) + mMgCl_2\cdot(-6.1) + mKCl\cdot(-0.16) \tag{1}$$

$$\Delta\delta^{18}O = mMgCl_2\cdot(1.11) + mCaCl_2\cdot(0.47) + mKCl\cdot(-0.16) \qquad (2)$$

where m is the molality, and $\Delta\delta^{2}H$ and $\Delta\delta^{18}O$ are the activity corrections, defined as the differences between the δ-values on the concentration scale and the activity scale.

The isotope evaporation lines were modelled following the method described by Gonfiantini (1986). It assumes that the isotopic composition of a water body undergoing evaporation varies with the decreasing fraction of the remaining water volume.

3.1.3. *Geochemical computations*

The software PHREEQC (Parkhurst and Appelo, 2013) was used to calculate the chemical evolution of evaporated waters, the required molalities for the isotopic activity corrections, the partial pressure of CO_2, and the mineral saturation indexes of gypsum, anhydrite, halite, calcite, and dolomite. Due to the high water mineralization, the computations were performed based on the PITZER database (Plummer et al., 1988), including Pitzer's thermodynamic equations (Pitzer and Kim, 1974; Pitzer and Mayorga, 1974, 1973) for high ionic-strength solutions.

4. Results

4.1. *Hydrodynamics*

During the control period, the flow rate in the Aguileras spring decreased from 40.5 l/s in May 2014 to 8.3 l/s in September 2017 (Fig. 2A), with an average value of 21.9 l/s. In Grande lake, the wetland stage dropped from 12.95 to 7.59 m during the same period. In both cases, a downward trend is caused by a decrease in annual rainfall, although staggered due to attenuation during precipitation periods. So, after the beginning of the rainy months, a stabilization (or rise in December 2016) of discharge rate and wetland stage was recorded, followed by a progressive descent. These variations are lagged and smoothed with respect to rainfall, especially in Grande lake (Fig. 2A). The combined evolution of both data series described a logarithmic regression (Fig. 2B). According to the function line, if the wetland stage would go below the elevation of the Aguileras spring (787 m asl), the value of the discharge rate would be nearly null.

4.2. *Water chemistry*

Table 1 summarises the hydrochemical results derived from water sampling in Los Hoyos area. Water presents a wide range of mineralization, from freshwaters to brines, and the EC values vary from 0.4 mS/cm (H3) to a maximum of 220 mS/cm (M2). The mean water temperature of groundwater (17–17.4 °C) is close to the mean atmospheric value (16.5 °C), with relatively low coefficients of variation. On the contrary, the Grande and Chica lakes show wider water temperature variabilities (Table 1).

Based on their hydrochemical composition, three different groups of water can be distinguished (Figs. 1 and 3). Firstly, the water sample from the Vizcaino wetland (H3), located at a higher altitude, shows calcium-bicarbonate facies. The biggest group is formed by calcium sulfate waters, including samples from the Aguileras spring (M1), the Grande and Chica wetlands (H1 and H2, respectively), and the Tunel leakage (R2). All those points locate in the western part of Los Hoyos area, between 780 and 790 m asl. Finally, samples from the Fuente Camacho I spring (M2) and the Fuente Camacho II leakage (R1), located at the east, below 750 m asl, are of sodium-chloride type.

The described hydrochemical pattern is also related to a progressive increment of the mineralization. Groundwater drained in the Fuente Camacho area (M2 and R1) has higher EC values and greater contents of Cl^-, Na^+, SO_4^{2-}, Ca^{2+} and K^+ (Table 1). The rise of EC towards lower altitudes is related to an increment in most major ions, particularly Cl^- and Na^+. These two species increase several orders of magnitude (Fig. 4A and B), reaching concentrations as big as 191 g/l of Cl^- and 115 g/l of Na^+ in M2 (Table 1). SO_4^{2-} and Ca^{2+} also rise towards lower heights, but the progression is less steep (Fig. 4C and D), and the maximum contents are 4.7 g/l and 2.9 g/l, respectively (Table 1). Finally, the evolutions of the alkalinity ($HCO_3^- + CO_3^{2-}$) and Mg^{2+} do not show any evident pattern (Fig. 4E and F).

To analyze the hydrochemical variability in Los Hoyos area and define the relationship degree between the different controlled parameters, a Principal Component Analysis (PCA) was carried out using the 10 variables included in Table 2 and a total number of 158 observations. The resulting correlation matrix shows a highly positive relationship ($R^2 > 0.9$) of EC with Cl^-, Na^+, K^+ and Ca^{2+}. EC is also positively correlated to a lesser extent with SO_4^{2-} ($R^2 = 0.884$), while it has a negative relationship with pH ($R^2 = -0.763$). In general terms, there are high correlations between most of the chemical species, except for Mg^{2+} and HCO_3^-, being the last one negatively related to pH ($R^2 = -0.818$).

The two main factors of the PCA explained 88.4% of the hydrochemical variability of the sample set (Fig. 5), although the first factor (F1) accounts for the majority of the statistical weight (71.51%). F1 is defined by EC and most of the chemical species considered: Cl^-, Na^+, K^+, Ca^{2+}, SO_4^{2-} and, to a lesser extent, NO_3^- (Fig. 5A). Except for the last one, all of them are linked to the dissolution of evaporite minerals forming the dominant lithologies presented in the media, and they gain concentration towards lower heights. F1 is also negatively related to pH,

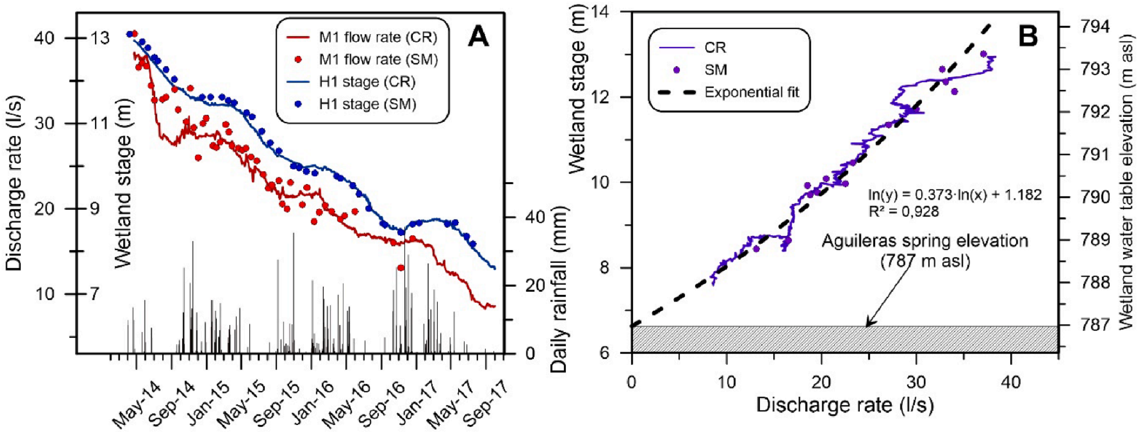

Fig. 2. (A) Evolution of the Aguileras spring discharge rate (M1) and the Grande lake stage (H1) between May 2014 and September 2017. (B) relationship between the records of the Grande lake stage and the discharge rate of the Aguileras spring, and logarithmic function between both datasets. CR, continuous record; SM, single measures.

Table 1

Main statistical descriptors (n, number of samples/measurements; min, minimum; max, maximum; mean, average; and cv, coefficient of variation in %) of physico-chemical data from surface water, groundwater, and rainwater in Los Hoyos area. Units: discharge rate/wetland stage (flow/w. stage) in l/s and cm, electrical conductivity (EC) in mS/cm, water temperature (Temp) in °C, pH in units of pH, major ions in mg/l, mineral saturation indexes (IS) are dimensionless, isotopic values in ‰ VSMOW (Vienna Standard Mean Ocean Water).

	CE	**T^a**	**pH**	**HCO_3^-**	**CO_3^{2-}**	**Cl^-**	**NO_3^-**	**SO_4^{2-}**	**Ca^{2+}**	**Mg^{2+}**	**Na^+**	**$\delta^{18}O$**	**δ^2H**	***d***	**SI_{CAL}**	**SI_{DOL}**	**SI_{ANH}**	**SI_{GYP}**	**SI_{HAL}**	**log pCO_2**
Springs																				
Aguileras (M1)																				
n	69	69	62	68	68	65	65	65	63	63	65	46	46	46	61	61	61	61	61	61
med	2.9	17.0	7.0	276	0	178	15	1,784	668	77	118	−5.8	−40.4	5.9	0.4	0.3	−0.4	0.1	−6.4	−1.6
máx	3.2	17.7	7.6	285	0	328	19	1,965	890	103	270	−3.0	−33.5	10.6	1.0	1.4	−0.3	0.1	−5.9	−1.0
mín	2.6	16.8	6.4	265	0	95	13	1,504	587	66	73	−6.7	−42.6	−9.6	−0.2	−1.0	−0.4	0.0	−6.8	−2.2
cv	6	1	3	1	0	32	7	6	5	6	32	−17	−6	n.d	n.d.	n.d.	5	44	4	14
Fuente Camacho I (M2)																				
n	37	36	31	37	37	37	35	37	37	37	37	8	3	3	37	37	37	37	37	37
med	215.5	17.3	6.1	240	0	173,493	21	4,219	1,966	143	107,341	−7.4	−41.5	21.3	1.0	1.4	0.2	0.4	0.2	−1.0
máx	220.0	21.3	6.7	261	0	191,334	87	4,734	2,903	212	115,617	−7.0	−40.5	23.6	1.6	2.8	0.5	0.6	0.4	−0.2
mín	210.0	13.2	5.4	207	0	155,785	1	3,676	1,729	55	86,801	−8.0	−43.5	19.7	0.5	0.0	0.0	0.2	−0.1	−1.6
cv	1	12	4	5	0	6	88	7	12	35	5	5	4	9	24	39	n.d.	30	n.d.	27
Leakages																				
Fuente Camacho II (R1)																				
n	63	63	55	62	62	60	55	60	62	62	62	8	3	3	58	58	58	58	58	58
med	193.8	17.4	6.2	318	0	130,883	29	3,706	1,815	165	82,478	−7.4	−42.2	21.7	0.6	0.5	−0.3	0.0	−0.4	−0.9
máx	203.0	18.8	7.1	362	0	150,962	85	4,408	2,531	254	109,693	−6.9	−40.9	22.6	1.4	2.5	0.1	0.3	0.0	−0.4
mín	174.1	16.4	5.7	283	0	112,763	3	3,078	1,627	65	63,720	−8.0	−43.3	20.8	0.2	−0.3	−0.4	−0.2	−0.7	−1.8
cv	3	4	3	6	0	6	74	8	10	28	9	7	3	4	23	n.d.	n.d.	n.d.	75	18
Tunel (R2)																				
23-02-2016	3.4	14.2	8.1	284	0	337	3.5	1,710	644	116	204	−3.2	−26.0	−0.7	1.4	2.3	−0.5	0.0	−5.8	−2.7
Wetlands																				
Grande lake (H1)																				
n	25	25	22	25	5	25	22	23	25	25	25	20	20	20	22	22	22	22	22	22
med	3.6	18.1	8.7	62	6	214	0.9	2,586	825	157	152	3.5	4.7	−23.2	1.2	2.1	−0.2	0.2	−6.2	−4.5
máx	4.2	28.7	9.6	90	8	278	2.7	3,124	946	197	192	6.4	18.4	−15.2	1.7	3.0	−0.1	0.3	−6.0	−3.0
mín	3.3	7.1	7.8	28	5	176	0.4	2,169	724	130	122	1.1	−7.3	−32.7	0.6	0.7	−0.4	0.1	−6.3	−8.1
cv	9	39	6	31	22	14	65	11	8	12	13	50	–	25	26	32	37	23	1	36
Chica lake (H2)																				
n	24	24	21	21	2	20	19	20	21	21	21	21	21	21	18	18	18	18	18	18
med	5.7	17.7	8.2	118	3.6	894	1.8	2,985	789	287	580	5.4	12.7	−30.1	0.9	1.6	−0.3	0.2	−5.1	−3.6
máx	8.1	30.8	9.2	165	5.8	1,476	3.9	4,337	997	464	920	9.2	30.8	−19.5	1.6	3.2	−0.1	0.3	−4.6	−2.0
mín	4.2	6.7	7.6	60	1.3	517	0.7	2,041	585	152	333	2.1	−3.0	−43.0	−0.1	−0.3	−0.5	0.0	−5.4	−9.9
cv	22	42	6	28	88	35	51	23	13	35	35	42	n.d	24	n.d.	n.d.	41	43	5	58
Vizcaíno (H3)																				
23-02-2016	0.4	17.4	7.7	227	0	5	0.2	12	77	2	3	−2.1	−9.5	7.6	0.4	−0.4	−2.8	−2.4	−9.4	−2.3
Rainwater																				
n				20	20	20	20	20	20	20	20	20	20	20	20	15	20	20	20	
med				11	0	2	1	5	6	0	2	−6.5	−39.2	12.8	−2.6	−5.7	−4.0	−3.8	−10.2	
máx				28	0	8	6	8	13	1	5	−3.1	−12.8	17.4	−1.7	−4.3	−3.4	−3.2	−8.9	
mín				3	0	0	0	4	1	0	0	−13.0	−95.3	8.2	−3.5	−7.5	−4.8	−4.5	−11.5	
cv				68	0	89	90	35	58	69	79	35	48	17	20	17	9.5	10	7.3	

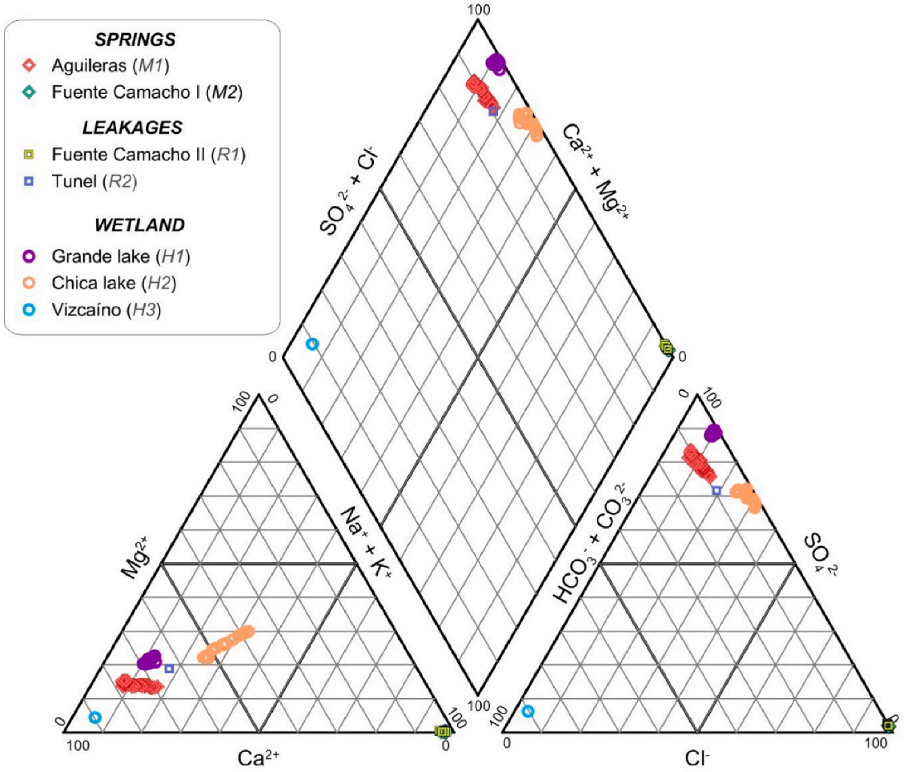

Fig. 3. Piper diagram showing the proportions of major ions dissolved in the water samples taken in Los Hoyos area.

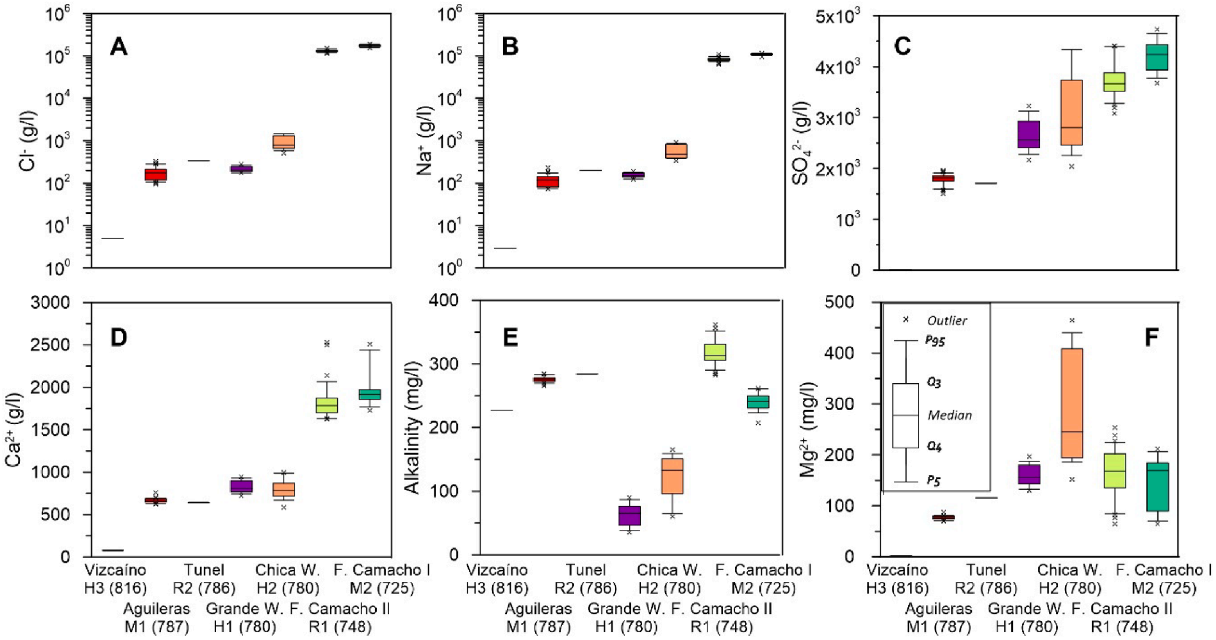

Fig. 4. Boxplots of Cl^-, Na^+, Alkalinity, Ca^+, SO_4^{2-}, and Mg^{2+} data of sampled points sorted by decreasing altitude (m asl), indicated in brackets.

with higher values in the wetland waters (Table 1). Thus, F1 reflects groundwater geochemical evolution, distinguishing between low mineralized water in equilibrium with the atmosphere (high pH) and those isolated from it (low pH) and with higher solute content. The second factor (F2, 16.93%) is defined by Mg^{2+} on its positive side and HCO_3^- on the negative one (Fig. 5A). Therefore, it is defined by those variables related to the calco-carbonic system that are not too influenced by gypsum dissolution as Ca^{2+} is. F2 could indicate the degree of equilibrium with the atmosphere or the evolution of surficial water.

Fig. 5B shows the observations (i.e. water samples) in the plane defined by F1 and F2. The brine waters of the Fuente Camacho sector (M2 and R1) are located on the positive side of the F1 axis. They are the samples with a higher dispersion according to the first factor. The rest of the observations are less mineralized, and they scatter in the left half of the plane in two groups along F2 (Fig. 5B). Samples from the Aguileras spring (M1), the Vizcaino wetland (H3), and the Tunnel leakage (R2) are

Table 2
Correlation matrix obtained from the PCA performed with the hydrochemical information collected in Los Hoyos area during the study period.

Variables	EC	HCO_3^{2-}	Cl^-	NO_3^-	SO_4^{2-}	Ca^{2+}	Mg^{2+}	Na^+	K^+	pH
EC	1									
HCO_3^{2-}	0.460	1								
Cl^-	0.992	0.425	1							
NO_3^-	0.722	0.470	0.717	1						
SO_4^{2-}	0.884	0.180	0.893	0.745	1					
Ca^{2+}	0.958	0.388	0.949	0.675	0.891	1				
Mg^{2+}	0.201	−0.293	0.185	0.316	0.537	0.270	1			
Na^+	0.991	0.417	0.995	0.695	0.880	0.948	0.169	1		
K^+	0.983	0.386	0.988	0.664	0.879	0.944	0.181	0.994	1	
pH	−0.763	−0.818	−0.757	−0.616	−0.517	−0.685	0.188	−0.757	−0.734	1

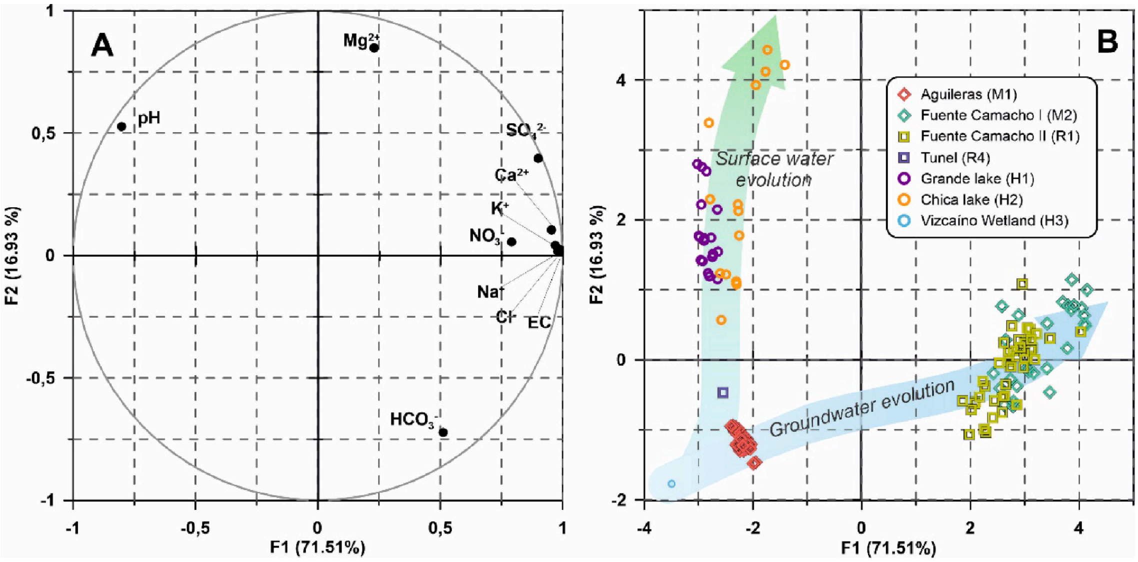

Fig. 5. Factors 1 and 2 obtained from the Principal Component Analysis (PCA) carried out with the main physicochemical parameters and major chemical components of the samples collected in Los Hoyos area during the study period. Plots of the variables (A) and statistical units (B).

in the lower left quadrant. They present low mineralization, relatively high content of HCO_3^-, and low pH. On the other hand, the samples of the Grande and Chica lakes (H1 and H2, respectively) scatter on the positive side of F2 (Fig. 5B), as they present high pH values, the lowest contents of HCO_3^- and the highest of Mg^{2+}. They have greater dispersion according to F2, particularly the Chica lake.

4.3. Characteristic molar relationship

In all the controlled points, the contents of Na^+ and Cl^- covariate (Fig. 6A) following the typical 1:1 stoichiometric line that defines the NaCl (halite) dissolution reaction. There is a clear difference between the Fuente Camacho area samples (M2 and R1) and the rest of the observations. The formers have Na^+ and Cl^- concentrations over 3500 meq/l, while the water of the other sampled points has less than 50 meq/l. Only the Vizcaino wetland (H3) sample presents a similar content of alkalinity and Ca^{2+} (Fig. 6B), indicating $CaCO_3$ dissolution as a source of both ions, while the rest present an apparent excess of Ca^{2+}. In Fig. 6C, showing the relationship between SO_4^{2-} and Ca^{2+}, springs and leakages samples plot on the 1:1 line that controls $CaSO_4$ dissolution reaction. Thus, it would be the primary geochemical process related to the origin of both ions in said waters. On the contrary, the samples from Grande (H1) and Chica (H2) lakes scatter below said line. That means that there is less Ca^{2+} than SO_4^{2-}, particularly in H2, which could be explained by calcite precipitation.

To interpret the geochemical evolution of Ca^{2+}, Mg^{2+}, and SO_4^{2-}, their contents are compared to Cl^- (Fig. 6 D-F). The brine samples of the Fuente Camacho area (M2 and R1) scatter separately from the rest, showing higher concentrations of SO_4^{2-} and, particularly, Ca^{2+}, and a positive relationship between those two species and Cl^-. On the contrary, Mg^{2+} values are in the range of the rest of the samples and no trend is observed (Fig. 6E). In the Grande and Chica lakes (H1 and H2, respectively), and to a lesser extent in Aguileras spring (M1), SO_4^{2-}, Ca^{2+}, and Mg^{2+} rise as Cl^- increases (Fig. 6 D-F). The covariation lines of Cl^--SO_4^{2-}, Cl^--Ca^{2+}, and Cl^--Mg^{2+} defined by the Grande and Chica lakes intersect the plotting space of the Aguileras spring, indicating a possible relationship. The linear regressions obtained from the molar relationships between Cl^- and Mg^{2+}, in both wetlands, and Cl^--SO_4^{2-}, in the Grande lake (H1), cross the origin (Fig. 6 E and F). That is, despite the contents of those ions vary, their molar relationships do not. Such a pattern is not observed for Cl^--Ca^{2+} covariation, nor for Cl^--SO_4^{2-} in the Chica lake (Fig. 6D and F), suggesting that Ca^{2+} and SO_4^{2-} are retired from the solution as Cl^- rises. Besides, the variations of the molar relationships of the Chica lake (H2) are more remarkable than in the Grande lake (H1), indicating that the processes causing such variability are more accentuated.

Fig. 6G show the Cl^- content *versus* the Cl^-/Br^- ratio (rCl/Br). The Fuente Camacho I spring (M2) presents rCl-/Br- values between 4500 and 7300, typically produced by halite dissolution (Alcalá and Custodio, 2008). Samples from the Aguileras spring (M1) and the Grande lake (H1) plot nearby a theoretical mixing/evolution line between recharge water and halite (or gypsum bearing halite) dissolution, although closer to the first type. The waters of the Tunnel (R2) and the Fuente Camacho II (R1) leakages, and, particularly, from the Chica lake (H2) plot below said evolution line, indicating an excess in Br^- typically produced by NaCl precipitation, often related to evaporation.

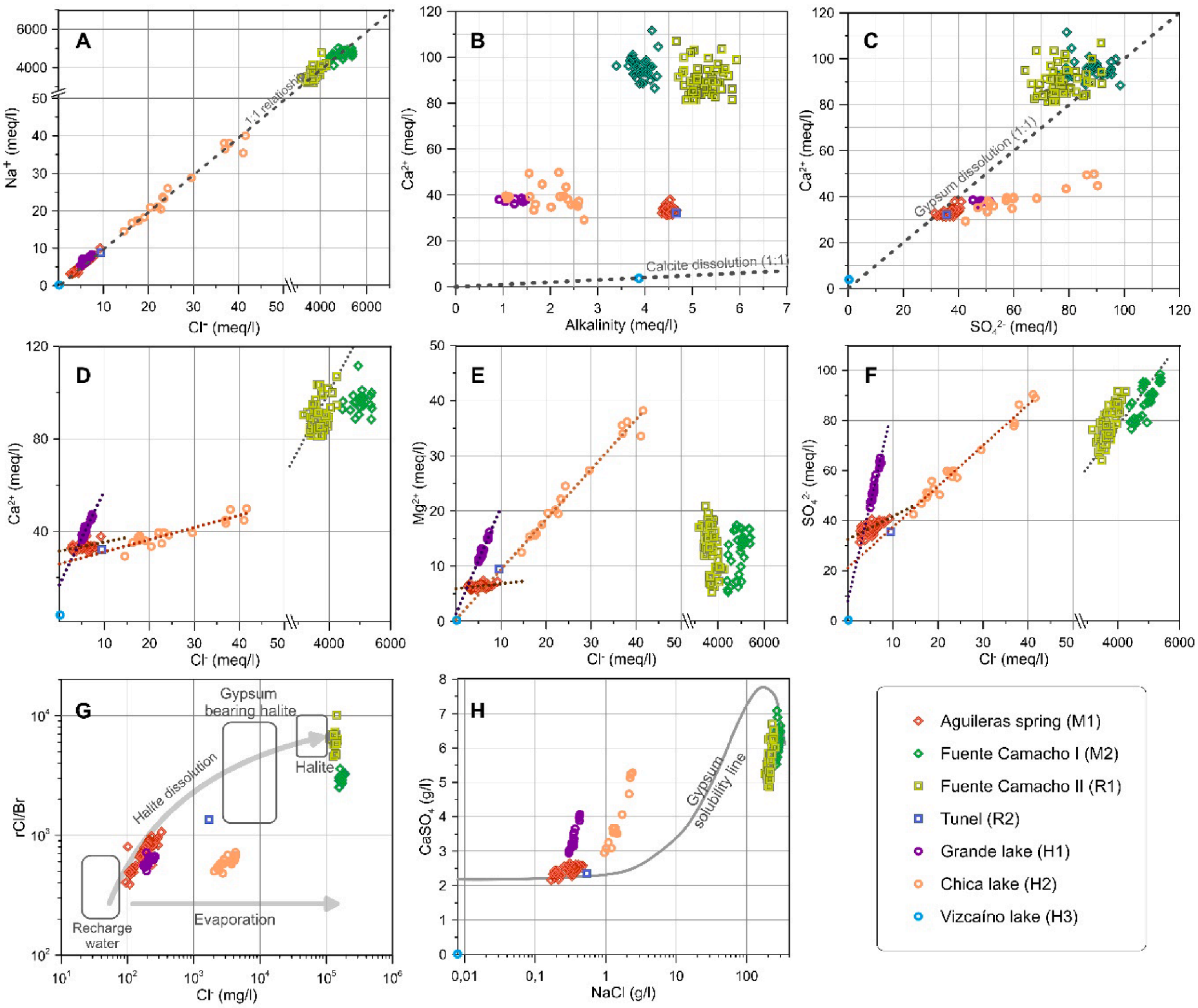

Fig. 6. Scatter biplots showing the relationships between (A) Cl^- and Na^+, (B) Alkalinity and Ca^{2+}, (C) SO_4^{2-} and Ca^{2+}, (D) Cl^- and Ca^{2+}, (E) Cl^- and Mg^{2+}, (F) Cl^- and SO_4^{2-}, (G) Cl^- and rCl/Br, and (H) NaCl and $CaSO_4$ concentrations.

4.4. *Saturation indexes*

The highest mineral saturation indexes (SI) correspond to dolomite (SI_{DOL}) and calcite (SI_{CAL}), whose respective maximum values are 3.2 in Chica lake and 1.7 in Grande lake (Table 1). The most negative saturation states have been determined for halite (SI_{HAL}), particularly in the waters of the Aguileras spring (M1) and the Vizcaino wetland (H3), which respectively reach minimum values of −6.8 and −8.7. Most water samples are undersaturated in halite and CO_2 (Table 1). However, in the Fuente Camacho area (M2 and R1), the maximum pCO2 values are close to saturation ($\log_{10}$ pCO2~0), and the SI_{HAL} even reach positive values, particularly in the Fuente Camacho I spring (M2). In contrast, wetland waters are undersaturated in CO_2 (Table 1), with mean values of log10 pCO_2 close to equilibrium with the atmosphere (−3.5; Appelo and Postma, 2005). Finally, concerning the saturation indexes of gypsum (SI_{GYP}) and anhydrite (SI_{ANH}), all the samples have values close to 0, except for the Vizcaíno wetland (H3), whose water is undersaturated in both minerals (Table 1).

Fig. 6H shows the contents of dissolved NaCl against $CaSO_4$, together with the line of evolution of $CaSO_4$ solubility as a function of the mass of NaCl dissolved in water at 20 °C and 1 atm pressure (Marshall and Slusher, 1966). The solubility of $CaSO_4$ increases slightly below 2.5 g NaCl/l in the range of the wetlands waters and brackish groundwater (H1 and R2). However, there is a marked increase in solubility to over 7.5 g $CaSO_4$/l for NaCl contents between approximately 100 and 250 g/l (Fig. 6H). That occurs due to the high ionic strength of saline waters that allows greater mineral dissolution rates, more accentuated in the case of gypsum and anhydrite (Zen, 1965). However, above 250 g NaCl/l, the solubility of $CaSO_4$ begins to drop significantly. Variations in the gypsum saturation state of water in Los Hoyos area are related to the halite dissolution state. Thus, in most samples, $CaSO_4$ concentrations are close to those that theoretically would be dissolved if the NaCl contents were equal to those determined (Fig. 6H). However, in the samples of the Grande (H1) and especially Chica wetlands (H2), there is an excess of $CaSO_4$ compared to what expected from the measured NaCl concentrations.

4.5. *Stable isotopes*

The mean values of $\delta^{18}O$ and δ^2H (Table 1) vary from −7.4‰ and −42.2‰, in the Fuente Camacho area (M2 and R1), to 5.4‰ and 12.7‰, in the Chica wetland (H2). Wetland waters show the most significant isotopic variability, with coefficients of variation in $\delta^{18}O$ of up to 50%, in Grande lake (H1). The excess in deuterium (*d*) ranges from −43‰, in the Chica lake (H1), to 23.6‰, in the Fuente Camacho I spring (M2). Waters from the Grande and Chica lakes always have negative values of *d* (Table 1), denoting an enrichment in heavy oxygen isotopes, often attributed to isotopic fractionation produced as a consequence of evaporation from a free sheet of water (Gonfiantini, 1986).

Fig. 7A shows the isotopic composition ($\delta^{18}O$ vs δ^2H) of rainwater,

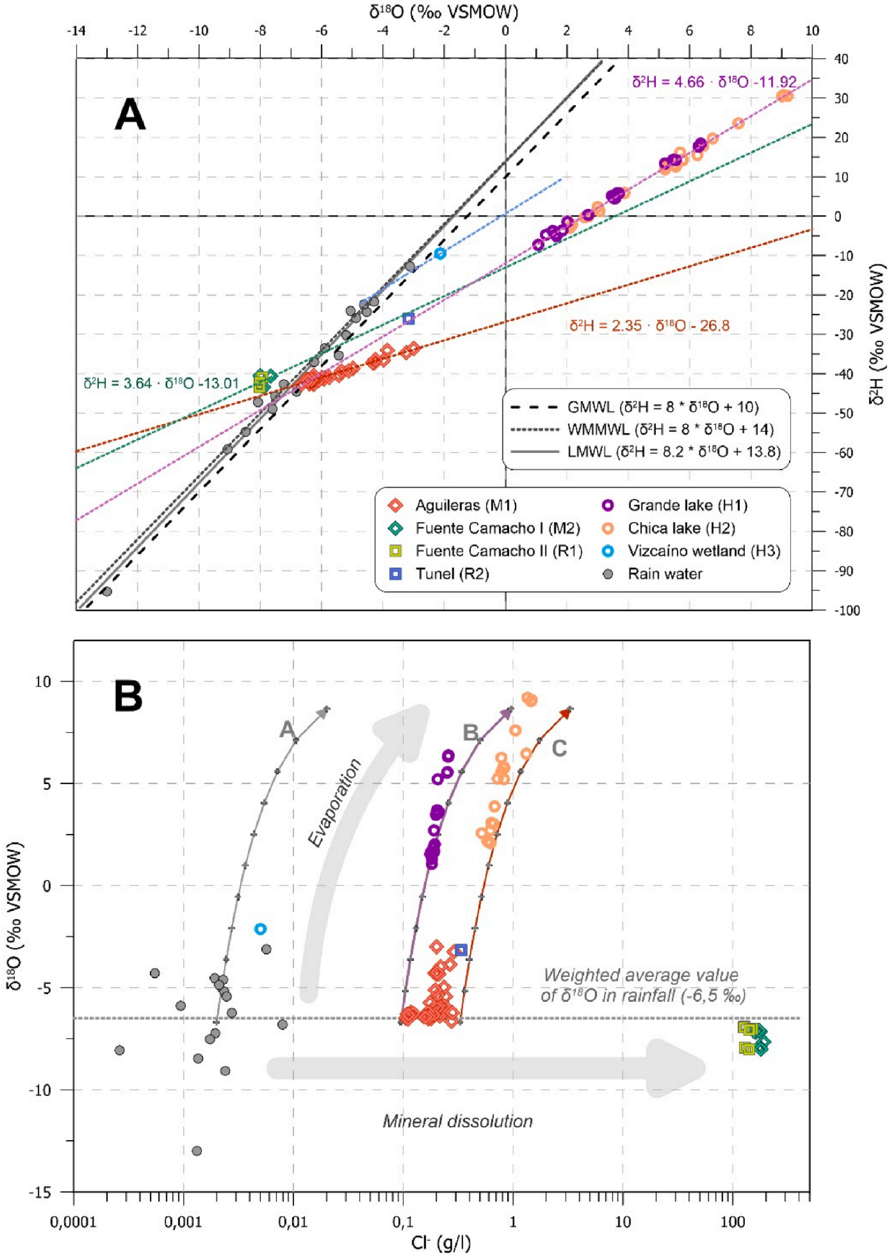

Fig. 7. Relationship between δ ^{18}O vs δ^{2}H from water samples (A). Global (GMWL), West Mediterranean (WMMWL), and Local (LMWL) Meteoric Water Lines and computed evaporation lines are displayed. δ ^{18}O vs Cl^{-} content (B). Computed isotopic and chemical evolution lines are also shown.

groundwater, and wetland water samples. Those from the Fuente Camacho area (M2 and R1) plot near the meteoric lines, while the rest of the observations scatter along evaporation lines. The waters from the Grande and Chica lakes align along a regression line with a 4.66 slope (Fig. 7A), characteristic of water bodies exposed to the atmosphere and, therefore, to evaporation processes (Mook, 2001). The water sample of the Vizcaíno wetland (H3) is placed in Fig. 7A near the Global Weather Meteoric Line (GWML), although slightly shifted to the right. Sampling in that wetland was performed in February 2016, during temporary flooding caused by a rainfall event characterized by δ^{18}O and δ^{2}H values of −4.30‰ and −21.74‰, respectively). The isotopic composition registered in the Vizcaino wetland could be explained by the evaporation of that rainwater, assuming an evaporation slope similar to that defined from the Grande and Chica lakes samples (Fig. 7A). The observations corresponding to the Aguileras spring (M1) distribute from δ^{18}O and δ^{2}H values close to the mean precipitation (−6.5‰ and −39.4‰, respectively; Table 1) towards heavier isotopic compositions, especially oxygen (Fig. 7A). The deviation of the samples from the meteoric lines reflects isotopic fractionation due to evaporation that, according to the slope calculated by linear fit (2.35), would have taken place in the subsurface rather than in free surface water (Geyh, 2001). Nonetheless, the most fractionated samples are those collected at the beginning of the study period, while subsequent observations have progressively varied their isotopic composition towards values closer to those of precipitation.

In Fig. 7B, the Cl^{-} contents of samples, which can be considered a conservative element (Appelo and Postma, 2005; Portugal et al., 2005), were plotted against their δ^{18}O isotopic values. Brine groundwater samples (M2 and R1) and most of the Aguileras spring ones (M1) scatter around a line with a δ^{18}O composition similar to the mean precipitation (−6.5−; Table 1) but with increasing mineralization (Cl^{-}). In contrast, the observations corresponding to the wetlands, the tunnel leakage (R2) and some of the Aguileras spring samples have δ^{18}O values markedly higher, indicating fractionation, but these show more moderate variations in mineralization (Fig. 7B). The theoretical chemical and isotopic evolution of rainwater (A) and two of the samples from the Aguileras spring (B and C) as its liquid fraction decreased due to evaporation was calculated applying the methodology proposed by Gonfiantini (1986). The Grande and Chica lakes (H1 and H2, respectively) samples scatter close to the evaporation lines simulated from the spring water (B and C), suggesting that the origin of the water stored in the wetlands could be related to groundwater inputs. Meanwhile, the evaporation of rainwater (A) could explain the mineralization observed in the Vizcaíno wetland (H3).

5. Discussion

5.1. Groundwater-wetland relationship

In general, the limnimetric variations recorded in Grande lake occurred in a lagged and attenuated manner with respect to precipitation events (Fig. 2A). In addition, the small rises in the water sheet that have happened after the rainfall events were of small magnitude compared to those observed on a weekly or monthly scale. All this evidences water input in the water balance of the wetland other than precipitation and runoff, as there are no surface water inputs after rainfall events. Thus, groundwater feeding to Grande lake is clear, as previous studies have already shown (CMA, 2005; Rodríguez-Rodríguez et al., 2006). On the other hand, the limnimetric decrease recorded during the hydrological years 2014/15, 2015/16, and 2016/17 accounts for up to 4150 mm. The accumulated evapotranspiration registered in a closed meteorological station (RAIF, 2021) for that period was 4574 mm, while the rainfall added 1254 mm. Therefore, if evaporation were the only wetland outflow, the theoretical descent would have been 3320 mm, even less assuming some surficial runoff and, particularly, groundwater inputs. Hence, groundwater outflow must also participate in the Grande lake water budget.

The proximity of the Aguileras spring to Grande lake (Fig. 1) and the lower elevation of the outlet could suggest a hydrogeological connection between them. However, the isotopic signatures of their waters are clearly different (Fig. 7A), as previous works have also shown (Gil-Márquez et al., 2016; Rodríguez-Rodríguez et al., 2007). While the Grande lake samples show a fractionation consistent with evaporation from free surface water (slope of 4.66), the spring waters follow a different evaporation trend (2.35) that is consistent with evaporation in the subsoil (Geyh, 2001). That kind of process could occur if recharge water is infiltrated slowly and diffusely, at the bottom of the dolines or in the rest of the recharge area. There, the soil and the epikarst could retain the water, which would be subject to evaporation before being incorporated into the saturated zone. According to Gil-Márquez et al. (2016), this process would have occurred predominantly in wet years (as in the one preceding the study period), explaining the more significant fractionation in the samples taken during the first year of observation. On the contrary, fast infiltration via karst forms would be predominant in dry years (as the study period), agreeing with an isotopic composition closer to rainfall.

Therefore, the isotopic information does not prove a direct hydrogeological connection from Grande lake to the Aguileras spring. On the contrary, the water drained by the outlet cannot come from the wetland, at least to a large extent. However, the positive relationship between the spring discharge rate and the wetland stage is remarkable (Fig. 2). The projection of the equation defining this association towards low values of the water stage would be related to a decrease in the spring flow. The discharge rate would reach 0 l/s if the wetland stage decreased to 6.5 m (Fig. 2B), placing the water table at 786.8 m asl, close to the spring elevation (787 m asl). Thus, there is a hydrodynamic relationship between Grande lake and the Aguileras spring, although it does not necessarily imply a direct dependence. There could be no direct hydraulic connection between the lake and the outlet, but instead, both drain the same hydrogeological system. In this way, local piezometric variations would affect both the discharge produced through the Aguileras spring and the limnimetric variations in Grande lake. The differences observed in the hydrodynamic responses of the spring and the wetland (Fig. 2A) could be partially caused by the store of water in the lake. The groundwater output could occur slower than the inputs and, therefore, the descends are delayed and enhanced in warmer months, when the evaporation rate is higher. In addition, the high heterogeneity of the media would determine the hydrogeological functioning of the spring and the wetlands. The predominance of low permeable clayey materials would condition the velocity of the lake inflows and outflows to be slow (medium-high inertia) and limit the underground connection between Grande lake and the Aguileras spring. On the other hand, the groundwater flow is inferred towards the NW from the water table sketch (Figs. 1 and 8), while the spring is westwards of the lake. Therefore, the hydraulic gradient would not be consistent with the wetland-spring direct connection without a preferential flow path produced by secondary porosity, which is not the case since the smooth discharge variations observed in the spring hydrograph (Fig. 2A). The water table sketch (Fig. 1) also indicates that groundwater flows beyond the edge of the diapiric structure. Therefore, neither the lake nor the spring must be the final destination of groundwater flows (Fig. 8). Thus, the gallery's construction from which the outlet emerges may have enhanced the discharge through it.

The joint interpretation of $\delta^{18}O$ values and Cl^- concentration of samples (Fig. 7B) permits a deeper understanding of the wetland-groundwater relationship. The samples from Grande and Chica lakes (H1 and H2, respectively) plot near the evaporation lines simulated from the spring waters (B and C in Fig. 7B). That suggests that the origin of the water stored in the wetlands could be related to groundwater inputs, which would have similar mineralization and isotopic composition to the spring samples. On the other hand, the evaporation of rainwater (A in Fig. 7B) could only explain the mineralization observed in the Vizcaíno wetland sample (H3). In Grande and Chica lakes, the geochemical evolution of Ca^{2+}, Mg^{2+} and SO_4^{2-} concerning Cl^- (Fig. 6D, E, and F, respectively) draw lines intersecting the spring samples space. Therefore, hydrogeochemistry reinforce the interpretation made based on the isotopic data. Thus, groundwater inputs are essential to maintain the water table in both the Grande and Chica lakes.

5.2. Hydrochemical processes and geochemical evolution of groundwater

The Na^+/Cl^- (Fig. 6A) and SO_4^{2-}/Ca^{2+} (Fig. 6C) molar relationships found in the water samples and their high contents of these ions (Table 1) evidence the dissolution of gypsum and halite as the most determinant hydrogeochemical processes affecting Los Hoyos area. The predominance of calcium sulfate and sodium chloride facies (Fig. 3) point out that way too. In addition, the existing lithological heterogeneity, together with the diversity of controlled water points, favours other types of physical and chemical processes that determine the geochemical evolution of water (Fig. 8).

Infiltration occurs through karst swallow holes and permeable rock outcrops and in the bottom of the dolines, where runoff water accumulates. The Vizcaino wetland (H3) is one of these dolines. The water retained in it could exemplify the chemical composition of the recharge waters, which are the starting point for the hydrogeochemical evolution in the system studied (Fig. 8). The water sample from the Vizcaíno wetland is poorly mineralized compared to those from the other water points sampled (Fig. 4 and Table 1). The elevated position of this wetland (816 m a.s.l.), above the piezometric surface (see Fig. 1), impedes it from receiving groundwater inputs. That suggests that its water's mineralization relates mainly to runoff inputs and rainfall accumulation, which also explains its short hydroperiod. Thus, water would have dissolved a small amount of solute mass on its path to the wetland, primarily calcite, as can be deduced from the HCO_3^-/Ca^{2+} molar ratio, close to the stoichiometric ratio (Fig. 6B).

After infiltrating, groundwater flows to lower elevations and

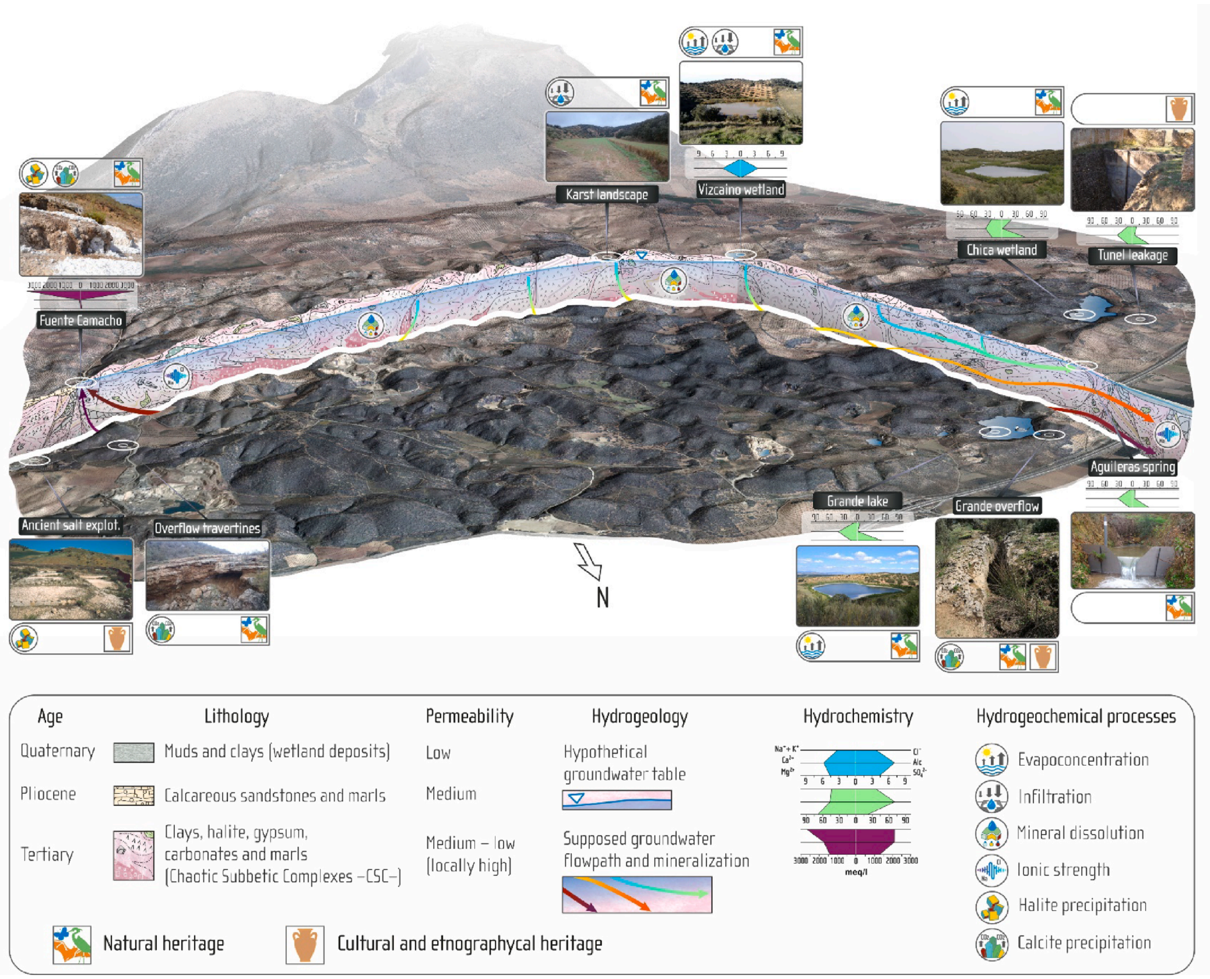

Fig. 8. Hydrogeological conceptual model of Los Hoyos area highlighting its hydrodiversity, the hydrological-hydrogeochemical processes and the related natural and cultural heritage.

dissolves the available minerals (Fig. 8): calcite, dolomite, gypsum, anhydrite and halite, among others. In the western sector of Los Hoyos area, at elevations between 780 and 787 m asl, most of the visible system's discharge occurs through the Aguileras spring (M1) and, to a lesser extent, via the Tunnel Leakage (R2); also by direct evaporation from the Grande and Chica lakes (H1 and H2, respectively). Groundwater in that sector, exemplified in the drainage of the Aguileras spring (M1), has reached saturation in the minerals mentioned above except for halite (Table 1).

Grande and Chica lakes would receive underground contributions with hydrochemical characteristics very similar to those of the Aguileras spring. However, evaporation from the water sheet of the wetlands would favour an increase in the concentration of dissolved ions (Fig. 8). This process would significantly impact the water of Chica lake due to its shallower depth, as evidenced by the rCl^{-}/Br^{-} ratios of its waters (Fig. 6G) and the higher increase in the concentration of the different ions in solution (Fig. 6). The rise of Mg^{2+} content due to evapoconcentration takes place in both wetlands (Fig. 6E) and produces a higher saturation in dolomite (Table 1). The amount of dissolved SO_4^{2-} and Ca^{2+} rises above the theoretical solubility values in both wetlands (Fig. 6H), leading to a slight rise in SI_{GYP} (Table 1). In Chica lake, where mineralization and the effect of evaporation are more remarkable, the Cl^{-} content increases to a greater extent than that of SO_4^{2-}; that is, while Cl^{-} reaches a fourfold increase in concentration, the second one only doubles it (Fig. 6F). That could be due to the removal of dissolved SO_4^{2-} as a consequence of gypsum precipitation. Finally, the Ca^{2+} concentration undergoes moderate increases compared to those of Cl^{-} (Fig. 6D) and SO_4^{2-} (Fig. 6E), in addition to an apparent deviation of the samples from the stoichiometric line defining the gypsum dissolution reaction. All this suggests that Ca^{2+} is being removed from the solution by calcite precipitation. Such a process is favoured by the high concentration of dissolved CaSO4, which enhances the calcite precipitation rate by the common ion effect (Wigley, 1973). In fact, travertine deposits are found by the overflowing threshold of the wetlands (Fig. 8). That would indicate that before the system was affected by any artificial drainage, the overflow of water from the lakes could have been frequent, favouring the degassing of the water, the precipitation of calcite and, consequently, the formation of travertine deposits. A similar process has been identified in the Ruidera lakes (Custodio, 2000). Another travertine formation appears by a capture doline on the NE border of the area (Figs. 1 and 8). The travertines developed by the open edge of the depression, similar to those found by Grande and Chica lakes, which could indicate that its origin is linked to a former wetland.

Groundwater flows towards the eastern edge of Los Hoyos area go to lower elevations (725–750 m asl) and, on their way, cross the saline core of the diapir structure. Recent research based on groundwater dating tracers (Gil-Márquez et al., 2020) has proved that part of the groundwater drained through this sector is older than the Aguilera spring water. It means that a portion of the drainage may come from recent infiltration, but most of the groundwater has undergone long-lasting underground flowpaths (Fig. 8). That allows for higher dissolution rates of gypsum, but especially of halite, as can be deduced from the increase in Ca^{2+}, SO_4^{2-}, Cl^{-} and Na^{+} concentrations as the discharge points locates at lower altitudes (Fig. 4). The drained waters in the

Fuente Camacho sector (M2 and R1) show rCl/Br values similar to those derived from halite (Fig. 6G), highlighting the high degree of geochemical evolution. The mineralization of the brine groundwater is associated with high ionic strengths, which increase the solubility of gypsum (Fig. 6H) and dissolve higher $Ca^{2+}$and SO_4^{2-} contents than those determined in the waters of the Grande and Chica lakes (Fig. 6C), which were already saturated with respect to this mineral (Table 1). The great amounts of dissolved Ca^{2+} and the presence of HCO_3^- favour the oversaturation respect calcite and, consequently, its precipitation, triggered by the common-ion effect with gypsum. The high values of SI_{HAL} also produce halite precipitation. However, halite deposits are washed out after rainy events while calcite remains, leading to travertine deposits linked to the brine outlets (Fig. 8).

All the hydrogeochemical processes described demonstrate the existence of hydrodiversity in Los Hoyos site, including waters of different chemical compositions as a consequence of the geodiversity (different lithologies, diverse karst landforms, etc.), but also of varying residence time within the media and flow-path length. Therefore, in Los Hoyos area, geodiversity conditions hydrodiversity, mainly from a chemical standpoint.

5.3. Hydrogeodiversity of Los Hoyos area as part of its natural and cultural heritage

A relevant functional implication of the geodiversity and particularly of the hydrodiversity in Los Hoyos area is the biodiversity of the ecosystems. Grande and Chica wetlands are among the deepest lakes in Andalusia (South Spain). They have regional ecological importance as they are the only lakes that present a mixing dynamic in the region (Moreira and Montes, 2005; Rodríguez-Rodríguez et al., 2001). These two wetlands host diverse species of flora and fauna, including some vulnerable or endangered species (Morales, 2009), such as *Zannichellia contorta* (García Murillo et al., 2018), *Fulica Cristata*, and *Aythya nyroca* (Birdlife International, 2021). For these reasons, among others, the Grande and Chica lakes are protected by the Andalusian Government as Natural Reserves, are considered a Natura2000 Special Protection Area (SPA) under the European Birds Directive, and are included in the Ramsar List of Wetlands of International Importance. However, the ecosystemic value of Los Hoyos goes beyond these two lakes with permanent or semipermanent hydroperiod. Most dolines can store surficial water for different time intervals and with diverse mineralization degrees. In addition, the Aguileras spring surrounding also presents characteristic biota. Consequently, the plant communities that develop in each of these features are different and give rise to diverse types of habitats (Morales, 2009; Ortega et al., 2002). Therefore, geodiversity and hydrodiversity conditions the biodiversity. Even more, all they are part of the natural and cultural heritage of the site.

According to Gray (2005), humankind should protect geodiversity against hazardous anthropogenic activities because of its intrinsic, cultural, aesthetic, economic, functional, and scientific values. Intrinsic value involves the ethical and philosophical dimensions of the relationships between nature and society and refers to the belief that something is valuable just for being what they are instead of the use that can be made of it (Nikitina, 2016). This value is often difficult to justify or understand. Still, if the relationship between society and the environment is assumed to be a determining factor, it should be noted that there have been human settlements nearby Los Hoyos area from Paleolithic times to the present (Toro and Ramos Lizana, 1988). Precisely, there is evidence of salt extraction from the brine outlets of Fuente Camacho, at least from the Copper Age, and more recent archaeological sites related to salt exploitation (Gómez Comino, 2011; Terán Manrique and Morgado, 2011). This is part of the cultural value of Los Hoyos (Fig. 8), which is directly related to the hydrogeochemical processes affecting groundwater and, therefore, contributing to the hydrodiversity of the area.

The aesthetic value of the abiotic nature in Los Hoyos is undeniable. The karst landscape developed over the outcrop is full of dolines and sinkholes, known locally as “hoyos”, from which it takes its name. Indeed, the geomorphological richness of this enclave led to its declaration as a site of geological interest (LIG) registered in the Andalusian inventory of geo-resources (CMAYOT, 2011). Nonetheless, the hydrogeological characteristics were also considered in such a definition. In addition, one of the items valued in the declaration of the Grande and Chica lakes as Ramsar sites was their contribution to landscape heterogeneity, as they represent water elements immersed in a relatively arid environment (Morales, 2009). The two wetlands are one of the main attractions of the hiking and biking trails in the area, which are environmentally friendly leisure activities that can indirectly provide an economic return for the surrounding communities. Other economic values of the geodiversity of the area are the gypsum and ophite (subvolcanic rocks) extraction for construction and the salt extracted from brine evaporation. While the exploitation of the two first types of mineral considerably impacts the landscape, the latter, directly related to the hydrodiversity, enhances the landscape variability with an original distinctive item: the white salt deposits over the dark coloured bedrock (Fig. 8). Finally, the use of water also provides economic returns. In the past, before the protection of the wetlands, the water of the Grande and Chica lake was channelled and used for diverse purposes, including the action of mills. Some remains of the infrastructures constructed to that end are still preserved, including the gates of the tunnel draining Chica lake (Fig. 8). Nowadays, a community of irrigators uses the groundwater drained through the Aguileras spring for agricultural production of nearby crops.

From the functional point of view, Los Hoyos is a well-developed gypsum karst system with numerous depression forms, where dissolution processes are still active due to the halokinetic elevation of the ground (Calaforra and Pulido-Bosch, 1999; Pezzi, 1977). Preferential infiltration forms (karst swallow-holes) are also abundant and, together with diffuse recharge through permeable outcrops and concentrated infiltration in doline beds, constitute the groundwater feeding of the system (Gil-Márquez et al., 2016). The aquifer discharge occurs in diverse forms (springs, wetlands, leakages) and with different mineralization, depending on the hydrogeochemical evolution of groundwater and, ultimately, the evapoconcentration of solutes (Fig. 8). This last process and the oversaturation of many mineral species in the brine groundwater of the Fuente Camacho outlets give rise to unique travertine deposits and halite precipitates (Fig. 8), being the latter nonpermanent. The travertine formations generally appear linked to carbonated water springs or waterfalls in fluvial curses where they often host wetlands (e.g., Collados-Lara et al., 2021; Florsheim et al., 2013). Although they may also form in palustrine environments (Alonso-Zarza and Wright, 2010), the appearance of travertines in the overflow area of wetlands, as observed in Grande and Chica lakes, is not well documented in the literature, except for some notable cases like the Ruidera lakes, in Spain (Custodio, 2000; Ordóñez et al., 2005) and the Plitvice Lakes, in Croatia (Chafetz et al., 1994).

All the geological, hydrogeological, and ecological processes in Los Hoyos area justify that the site has served, for several decades now, in teaching activities of different courses taken at near universities of Malaga and Granada. The visit to the area as part of the programme of several international scientific conferences (V International Symposium of Karst, 2014; 46th IAH Congress, 2019) also evidences the scientific and informative value of the area.

In addition to these basic five types of values of geodiversity, Doyle and Benneth (1998) also consider the information value of abiotic nature by analogy with the genetic diversity of biota. It refers to the fact that each geological object contains unique information, and at present, only a fraction of it is currently known or understood (Nikitina, 2016). In that sense, Los Hoyos hydrodiversity is a consequence of the different water-rock interaction processes occurring in the media, some of which have been explained in this work. However, the progress of analytical determination techniques and hydrogeological knowledge, in general, may allow the identification of many others in the future and, for that reason, such a particular place like Los Hoyos area must be protected.

In light of all the above, the natural and cultural heritage of Los Hoyos is largely due to its geodiversity. Most of it is linked to hydrodiversity, understood as the natural range of hydrological manifestations that exist in the area, including not only the typology but also its natural variations (hydrochemistry and hydrodynamics) and all the hydrogeological processes causing them. According to (Simić et al., 2012), the protection of water phenomena cannot be the subject of interest only for individuals or the scientific community. Instead, the total commitment of communities is necessary to protect the hydrological heritage. In that sense, it is essential to advance in the definition and promotion of geological diversity, particularly hydrodiversity, so that society comes to value the hydrological heritage as it currently respects biological diversity.

6. Conclusions

This work evidences how geodiversity conditions the hydrodiversity of a place and, ultimately, its natural and cultural heritage. In that sense, the existence of a wide variety of water manifestations (springs, wetlands, leakages) of diverse mineralization and hydrodynamic is essential for establishing rich flora and fauna (biodiversity). Moreover, the development of human activities, from ancient times to the present day, is conditioned by the availability of water, in different forms and with diverse hydrochemical characteristics.

Los Hoyos area (S Spain) is presented as an illustrative example of hydrodiversity. Chemical and isotopic tools were used to analyse the characteristics of its wetlands, springs, and leakages. The interpretation of the results has enhanced the hydrogeological knowledge of the system, particularly regarding its hydrogeochemical processes. Thus, the geochemical evolution of water in the site depends (1) on the availability of the minerals that determine the hydrochemical facies, (2) the residence time of groundwater flows, (3) the free-surface evaporation in wetland waters, (4) the common-ion effect, and (5) the high ionic strength of groundwater.

Three main groups of waters have been identified according to their chemical and isotopic composition. Freshwaters (seasonal recharge wetlands), with calcium carbonate water, brackish waters (west discharge area, including permanent wetlands), with calcium sulphate waters, and brine water of sodium chloride type (east discharge area). The progressive mineral dissolution explains this distribution along with the groundwater flow. A longer water-rock interaction and the presence of halite at depth are the reason for the brine groundwater in the eastern sector, characterized by high ionic strength, which enhances gypsum dissolution. Meanwhile, the evapoconcentration of water in the wetlands favours mineral precipitation, particularly calcite, which is triggered by the common-ion effect with gypsum. This leads to the formation of travertine deposits, especially in the overflow areas of wetlands but also nearby the brine outlets.

The diversity of water elements and hydrogeochemical processes in the area (hydrodiversity) is directly linked to the wide range of existing lithologies, the characteristic karst geomorphology of the site (geodiversity), and its inner structure. All that combined conditions the hydrogeological functioning of Los Hoyos area and, ultimately, its great hydrodiversity. Even more, hydrodiversity is vital for the rich biodiversity of the area, mainly concentrated in the wetland environment, and its cultural heritage, which is closely related to the use of water and salt extraction from brine groundwater (since the Paleolithic). Thus, the protection of all those natural and cultural features is not possible without preserving the hydrological heritage of the area; not only the water manifestations themselves (springs, wetlands) but all the elements involved in their natural dynamics (recharge areas, flow paths, groundwater-surface water relationship, among others).

In that sense, it is necessary to advance in the definition and promotion of the hydrological heritage so that public administration and society, in general, appreciate the value of hydrodiversity. Advancing in the definition of Gil-Márquez et al., 2022, hydrodiversity could be described as the set of natural water manifestations that exist in an area, continental or marine, surface or groundwater. The basic criteria for its qualitative assessment would be related to its typology (genesis), its natural variations (hydrochemistry, hydrodynamics, freeze/thaw) and the hydro(geo)logical processes that cause them. In the case of groundwater, hydrodiversity is fundamentally motivated by the diversity of origins and their relationship with the dominant lithologies, hydrochemical facies and residence time in the subsoil. Analogous to geodiversity, hydrodiversity offers society a series of services related to its intrinsic, cultural, aesthetic, economic, functional, and scientific values.

7. Research perspectives and recommendations

This work has shown how an in-depth analysis of hydrogeological processes helps to explain the diversity of groundwater manifestations, and it highlights the importance of such processes on the ecosystem and social services. However, following the definition of hydrodiversity given in the previous section, it would be necessary to carry out this type of analysis in surface and coastal waters too, or even in other groundwater systems, in order to assess how hydro-bio-geo-physico-chemical processes intervene in their variability and, ultimately, on the ecosystem and social services.

To advance in the definition and acceptance of the term hydrodiversity, it is necessary to carry out interdisciplinary work involving hydrogeologists, environmentalists, ecologists, geographers, and any other scientific field that studies the hydrosphere but also researches from the social sciences (e.g., historians, economist). In this way, the necessary consensus to describe hydrodiversity holistically could be reached. Only by explaining the diversity of hydrological heritage and the benefits it brings us can society be made aware of the importance of its protection.

At present, some countries directly or indirectly preserve hydrodiversity by protecting certain hydrological elements through different legal figures. For example, some hydrological features of great beauty, such as springs or waterfalls, are protected as natural or national monuments. A particular case are wetlands, which are often protected natural areas due to the biodiversity they can harbour. In other cases, water objects are included in the declaration of some UNESCO global geoparks. Some authors agree with the idea that the hydrological features are geological heritage, and thus they may be protected as such (Brilha et al., 2018). Others believe that unique water elements can be a distraction from valuing other purely geological aspects in geoparks and, therefore, another conservation figure should exist for the protection of the hydrological heritage (Ruban, 2019).

In any case, before proposing hydrodiversity protection figures, it will be necessary to assess the value of water elements in every region. Thus, it is essential to have proposals for the classification and identification of hydrological heritage, such as that of Simić et al. (2014) and to elaborate inventories of points of hydrological interest, such as those that the Geological Spanish Survey prepared for some regional public administrations. This, together with scientific studies that allow us to reach deeper insights into the hydrological processes of a region and their ecological and socio-cultural implications, will make it possible to manage and preserve hydrodiversity for future generations.

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