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Articles

Hydrogeochemical and isotopic processes that control the chemical variability of the waters in a sector of the Región Carbonífera Aquifer, Coahuila

Procesos hidrogeoquímicos e isotópicos que controlan la variabilidad química en un sector del Acuífero Región Carbonífera, Coahuila

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Abstract

The Region Carbonifera Aquifer is one of the 28 existing hydrogeological systems in the State of Coahuila and one of those that suffer the most pressure due to mining activity and overexploitation. This study is framed in the western portion of the aquifer, encompassing the Ejido Morelos and the Colonia El Nacimiento, both in the Melchor Muzquiz municipality. In this area, 14 sites were sampled (4 wells, 6 norias, 2 springs, and 2 sections of the Sabinas River) with the aim to characterize these water bodies from the hydrogeochemical and isotopic point of view, thus obtaining a conceptual model of this sector. The results obtained show the chemical variability of the sampled waters, classifying waters as bicarbonate-calcium, bicarbonate-sodium, sulfated-calcium, and chloride-sodium, which supports the occurrence of various processes that affect the quality of the resource, among them the salinity. From the isotopic point of view, a meteoric origin is proposed for most water bodies under study, whose recharge zone comes from Santa Rosa mountains. However, surface evaporation, the dissolution of salts, and ionic exchange processes generate chemical variability and isotopic fractionation, which conditions heavier isotopic signatures and more enriched and saline waters. These elements are represented in a conceptual hydrochemical and isotopic model, which integrates both interpretations and shows the hydrogeological complexity of the region in terms of the quality of the resource.

Keywords: Hydrochemistry, stable isotopes, Región Carbonífera Aquifer.

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Resumen

El Acuífero Región Carbonífera es uno de los 28 sistemas hidrogeológicos existentes en el estado de Coahuila, y también de los que más presiones sufre debido a la actividad minera y la sobreexplotación. Este estudio se enmarca en la porción oeste de dicho acuífero, abarcando el Ejido Morelos y la Colonia El Nacimiento, ambos en el municipio Melchor Múzquiz. En dicha área se tomaron muestras en 14 sitios (4 pozos, 6 norias, 2 manantiales y 2 secciones del río Sabinas), con el objetivo de caracterizar desde el punto de vista hidrogeoquímico e isotópico estos cuerpos hídricos y así obtener un modelo conceptual de dicho sector. Los resultados obtenidos muestran la variabilidad química de las aguas muestreadas, clasificándose como aguas de tipo bicarbonatada-cálcica, bicarbonatada-sódica, sulfatada-cálcica y clorurada-sódica, lo cual sugiere la ocurrencia de variados procesos que afectan la calidad del recurso, entre ellos la salinidad. Desde el punto de vista isotópico se propone un origen meteórico para la mayoría de los cuerpos hídricos en estudio, cuya zona de recarga proviene de la sierra de Santa Rosa. Sin embargo, procesos como la evaporación superficial, la disolución de sales, y el intercambio iónico generan variabilidad química y fraccionamiento isotópico, lo que condiciona firmas isotópicas más pesadas, y aguas más enriquecidas y salinas. Estos elementos se representan en un modelo geohidrológico e isotópico conceptual, que integra ambas interpretaciones y muestra la complejidad hidrogeológica de la región en términos de calidad del recurso.

Palabras clave: hidroquímica, isótopos estables, acuífero Región Carbonífera.

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Introduction

Salinity in Mexican aquifers has been a worrying issue in recent decades. According to Raynal-Gutiérrez (2020), 32 aquifers with salinity problems have been identified in Mexico, mainly in arid and semi-arid climates where low rainfall and high evaporation rates prevail. In addition, marine intrusion problems are reported in at least 18 coastal aquifers, most of them in the Baja California Peninsula and in Sonora (Torres-Martínez, Mahlknecht, Hernández-Antonio, & Mora, 2017; Raynal-Gutiérrez, 2020).

In other parts of the country, salinity has increased due to shallow groundwater levels, dissolution of evaporite minerals, the presence of connate or fossil waters, and the overexploitation of water resources (Wurl, Gamez, Ivanova, Lamadrid, & Hernandez-Morales, 2018; Gilabert-Alarcón *et al*., 2018). In addition, the effects of climate change indicate future problems in the availability and quality of water resources, as reported by Hassani, Azapagic and Shokri (2021) in their global predictive model.

In the State of Coahuila, the hydrogeological resources are distributed in 28 aquifers covering different climates, geological-structural conditions and, exploitation pressures. In fact, at the state level, data

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compiled in 2018 shows that a not inconsiderable 32% of aquifers suffer from salinity phenomena while seven aquifers show overexploitation (Conagua, 2020). The Region Carbonifera Aquifer is not exempt from the effects of salinization due to complex geology and the predominance of an extreme arid/semi-arid climate. Throughout the aquifer, research related to the chemical composition and water quality of the water bodies that comprise it is limited. The National Water Commission (Conagua) is the one that has studied the aquifer with some degree of detail based on its report "Update of Water Availability in the Region Carbonifera Aquifer, state of Coahuila" (Conagua, 2020) where it details the most relevant aspects of this system in terms of geology, geomorphology, hydrogeological characteristics and calculations of water availability and balance. A more detailed study is presented by Batista-Cruz *et al*. (2022), who studied 155 samples of water taken from wells, wells, springs, and surface sources. The study details important aspects of water quality based on NOM-127-SSA1-1994 (DOF, 2000) for consumption purposes and, in turn, shows the complex situation of the aquifer, whose quality is questionable for agricultural activities. However, due to the chemical variability observed by these authors, hydrogeochemical and isotopic studies of small areas within the aquifer can help to understand better what processes deteriorate the quality of the water resource and, in this sense, how to deal with it in municipalities and ejidos (i.e., Mexican term to call rural communities) the problem of water supply for various purposes.

On the other hand, isotopic studies (i.e., stable isotopes of oxygen and deuterium) are practically null, with only information in the nearby

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Cuatrociénegas and El Hundido aquifers (Johannesson, Alejandra, & Kilroy, 2004; Felstead, Leng, Metcalfe, & Gonzalez, 2015). The synergy between chemistry and isotopy allows for identifying dissimilar and complex hydrogeological and environmental processes and understanding those causes that can modify the pristine chemical composition of a given water body and its origin.

The goal of this study is to characterize, from the hydrogeochemical and isotopic point of view, the surface and groundwaters resources of the Ejido Morelos and Colonia El Nacimiento in the municipality of Melchor Múzquiz, Coahuila, to identify the geohydrogeological and isotopic processes that they undergo groundwaters in its movement through the natural environment and, in turn, with them, to develop a conceptual model of the origin of water in this sector of the Region Carbonifera Aquifer.

Study area

The study area focuses on Ejido Morelos and Colonia El Nacimiento, two contiguous zones that cover a total area of 521 km^2, located in the municipality of Melchor Múzquiz and the western portion of the ARC (Figure 1). The aquifer, classified as free type, is made up in its upper part by alluvial sediments and polymictic conglomerates of the Sabinas Formation. In its intermediate part is the Olmos Formation, and at greater depths, the calcareous rocks, fundamentally from the Aurora and Cupido Formations of the Lower Cretaceous, constitute the aquifer unit that can

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present confinement conditions because it is overlain by clayey rocks such as shale and siltstone (SGM, 2003; Conagua, 2020).

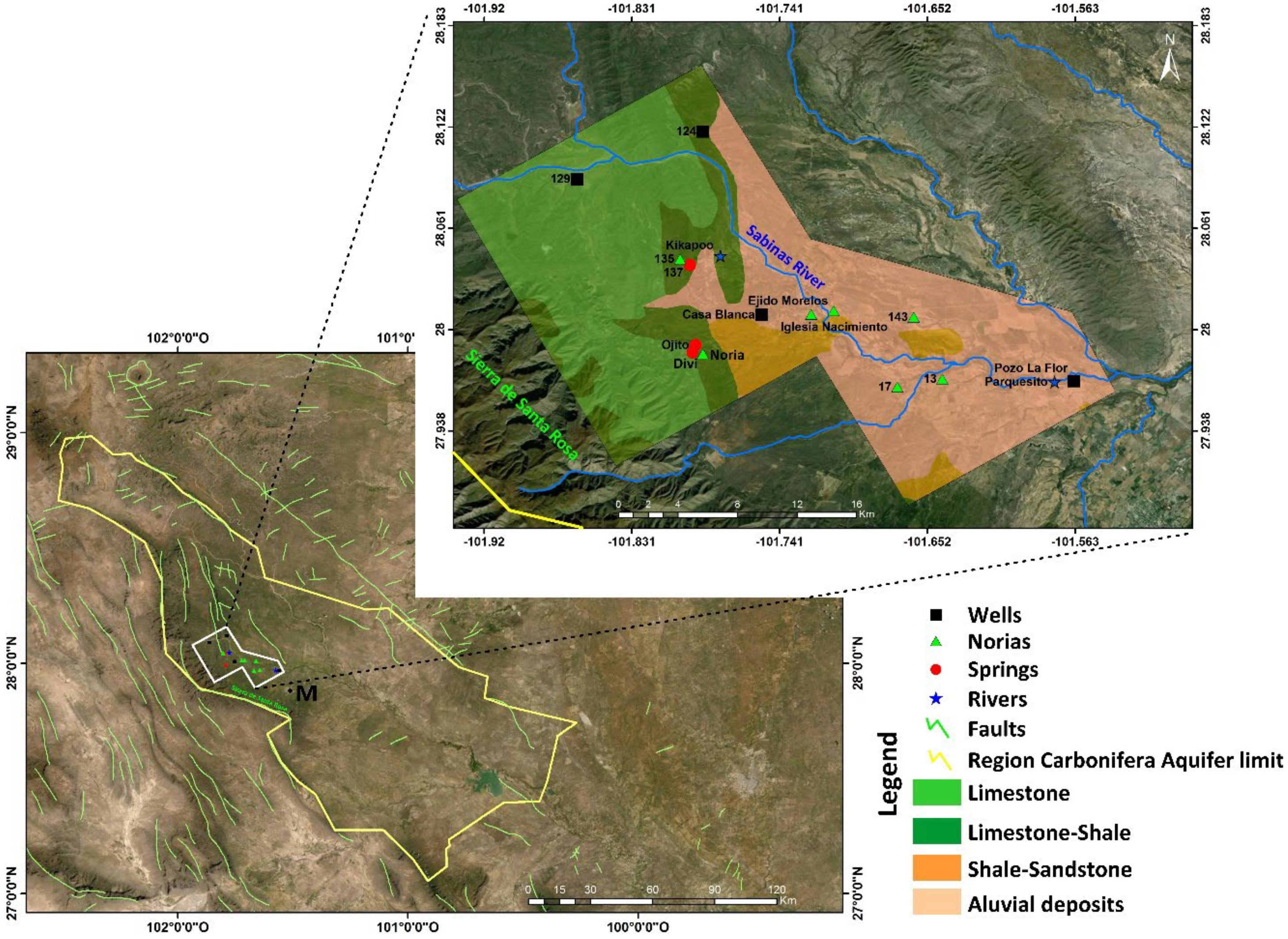

Figure 1. Geographical location map of the study area within the limits of the Region Carbonifera Aquifer in the State of Coahuila. The geological base was taken from SGM (2003). On the map: M-Melchor Múzquiz town.

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Within the aquifer's limits, the static level's depth varies from 5 to 30 meters. However, this depth can reach up to 50 meters in some wells drilled in limestone located in Sierra de Santa Rosa.

Surface hydrology is characterized by intermittent and ephemeral channels, with very few permanent channels, as in the case of the Sabinas River, which passes through the study area and whose isotopic data is presented in this study.

Materials and methods

Sampling and analytical techniques

Sampling work was carried out at 14 sites (four wells, six norias, two springs, and two sections of the Sabinas River) (Table 1). At each location, the coordinates, altitude, and the physical-chemical parameters (e.g., pH, conductivity, and total dissolved solids) were obtained (Table 1) with the help of a GPS Garmin and HANNA HI98194 multiparameter equipment respectively. At the end of each measurement, the equipment was washed with deionized water to avoid measurement errors and for its own care and good use. In the case of chemical sampling of major elements (e.g., Cl^-, SO_4^{2-}, HCO_3^-, Na^+, Ca^{2+}, K^+, Mg^{2+}), 500 milliliters of water samples were collected in previously washed High-Density Polyethylene (HDPE) bottles in a nitric acid solution. Additionally, the water samples for cations were acidified in the field with HNO_3 to pH = 2. For the case of

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the isotopic analyzes ($\delta^{18}O$ and δD), the water samples were extracted with a 50 ml syringe attached to a sterile conical VWR filter, with a diameter of 25 mm and a pore size of 0.45 micrometers. All samples were taken in duplicate and refrigerated in airtight coolers at a temperature between 1 and 5 degrees Celsius to avoid any type of contamination based on the works of Nollet (2000).

Table 1. Physical-chemical data of the water samples under study.

Samples	**Type**	**pH**	**TDS (ppm)**	**E.C (µS/cm)**	**HCO_3^- (mg/l)**	**SO_4^{2-} (mg/l)**	**Cl^- (mg/l)**	**Na^+ (mg/l)**	**K^+ (mg/l)**	**Ca^{2+} (mg/l)**	**Mg^{2+} (mg/l)**	**Error (%)**
Casa Blanca	Well	8.08	1579	3160	538	24.2	285.4	307	1.31	22.3	2.4	8.34
129	Well	8.3	385	460	237.9	21.5	12.3	3.7	0.4	81	12.2	-5.28
124	Well	7.8	1319	4770	2446.1	20.6	397.6	1244	5.1	37	18.5	-5.38
Pozo La Flor	Well	7.87	855	1711	476	71.2	26	188	1.23	12.3	2.5	5.19
143	Noria	7.7	451	540	305	11	10.01	3.7	0.4	101	8.8	-3.72
135	Noria	7.6	565	650	353.8	31.2	14.2	6.2	1.6	140	3.6	-5.16
13	Noria	7.9	176	320	109.8	11	10.7	4.4	1.6	32	6.4	-0.54
17	Noria	7.6	472	730	79.3	194.3	60.4	46	3.9	74	14.4	0.51
Ejido Morelos	Noria	6.69	895	1790	345	38.2	81.9	57.9	1.58	127	10.1	-5.23
Iglesia Nacimiento	Noria	7.52	2146	4305	499	9.6	474.4	383	1.8	20.6	3.4	9.42
Parquecito	River	6.79	551	1103	351	42.2	7.8	4.7	0.73	117	7.9	1.03
Kikapoo	River	8.03	383	766	185	53.7	9.7	10.1	0.99	69	10.8	-4.04
Ojito	Spring	7.33	589	1177	409	24.9	11.6	9.71	2.63	118	0.96	7.79
137	Spring	7.8	357	450	231.8	20.6	14.2	2.3	0.4	81	7	-1.07

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The physical-chemical analyzes at a laboratory scale were carried out in the Laboratory of the Faculty of Chemistry of the Autonomous University of Coahuila (UADEC), using various volumetric and analytical methods supported by the corresponding Mexican standards. In the case of Ca^{2+} and Mg^{2+} cations, their concentrations were calculated by multiplying the temporary hardness, previously calculated by titration with EDTA 0.2N (SCFI, 2001c) by the factors 0.4 and, 0.24 respectively. On the other hand, the concentrations of Na^+ and K^+ were obtained by flame atomic absorption spectrophotometry (AA240FS Varian) using calibration curves with five standards of 0.4 to 1.2 mg/l and 0.3 to 1.5 mg/l respectively, from certified standards of 1 000 mg/l (SCFI, 2001b). The concentrations of the anions HCO_3^-, SO_4^{2-}, and Cl^- were acquired by titration with H_2SO_4 at 0.02N (SCFI, 2001a), $BaCl_2$ at 0.1N (SCFI, 2014) and $AgNO_3$ at 0.01N (SCFI, 2001d), respectively.

The isotopic analyzes were performed with the Picarro L2130-I water isotopic analyzer owned by the Ensenada Baja California Center for Scientific Research and Higher Education. The equipment, based on the "Cavity Ring-Down Spectroscopy" (CRDS) technology, measures the isotopic ratio of δD, $\delta^{18}O$ with high precision (low errors less than 0.1 ‰) (Picarro Inc., 2008). The filtered samples without added acids are contained in small vials (up to 200 µl) and placed in an autosampler of the equipment, together with deionized water and the respective internal standards Pi-Z ($\delta^{18}O$ = 0.3 ‰; δD = 1.8 ‰), Pi -M ($\delta^{18}O$ = -20.6 ‰; δD = -159.0 ‰) and Pi-D ($\delta^{18}O$ = -29.6 ‰; δD = -235.0 ‰) and VSMOW2 certified international standards ($\delta^{18}O$ = 0 ‰; δD = 0 ‰), GISP ($\delta^{18}O$ = -24.7 ‰; δD = -189.9 ‰) and SLAP ($\delta^{18}O$ = -55.5 ‰;

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δD = -428.0 ‰). In each analysis, 10 measurements are obtained, and the first 5 ignored to eliminate the memory effect. Finally, the last 5 are considered for correcting the raw data based on international and internal standards by the linear regression method.

The reproducibility of the analyzes for $\delta^{18}O$ and δD was around ±0.1 ‰ and ±0.4 ‰, respectively. These data are reported in comparison with the Vienna Standard Mean Ocean Water 2 (V-SMOW2), while the results are reported in δ notation as deviations from isotopic ratios in parts per thousand (‰) according to Craig's criteria (Craig, 1961).

Results

The 14 samples under study present physical-chemical differences (Table 1), which presupposes the occurrence of various hydrochemical processes in said sites. The balance error calculated from the sum of anions and cations in meq/L yielded values less than 10%, which indicates optimal data quality (Appelo & Postma, 1994).

In detail, except for well 129, all the samples from the wells show a higher concentration of total dissolved solids (TDS) and in electrical conductivity (E.C.) (Figure 2A). In detail, the Casa Blanca (1 579 ppm) and 124 (1 319 ppm) wells and the Iglesia Nacimiento well (2 146 ppm) show concentrations of TDS above the maximum permissible value stipulated by the Official Mexican Standard NOM-127-SSA1-1994 (Figure 2A; TDS-1 000 ppm), therefore they present poor water quality for human

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consumption purposes. The rest of the wells, surface sources, and springs show TDS values below 895 ppm and E.C below 1177 µS/cm.

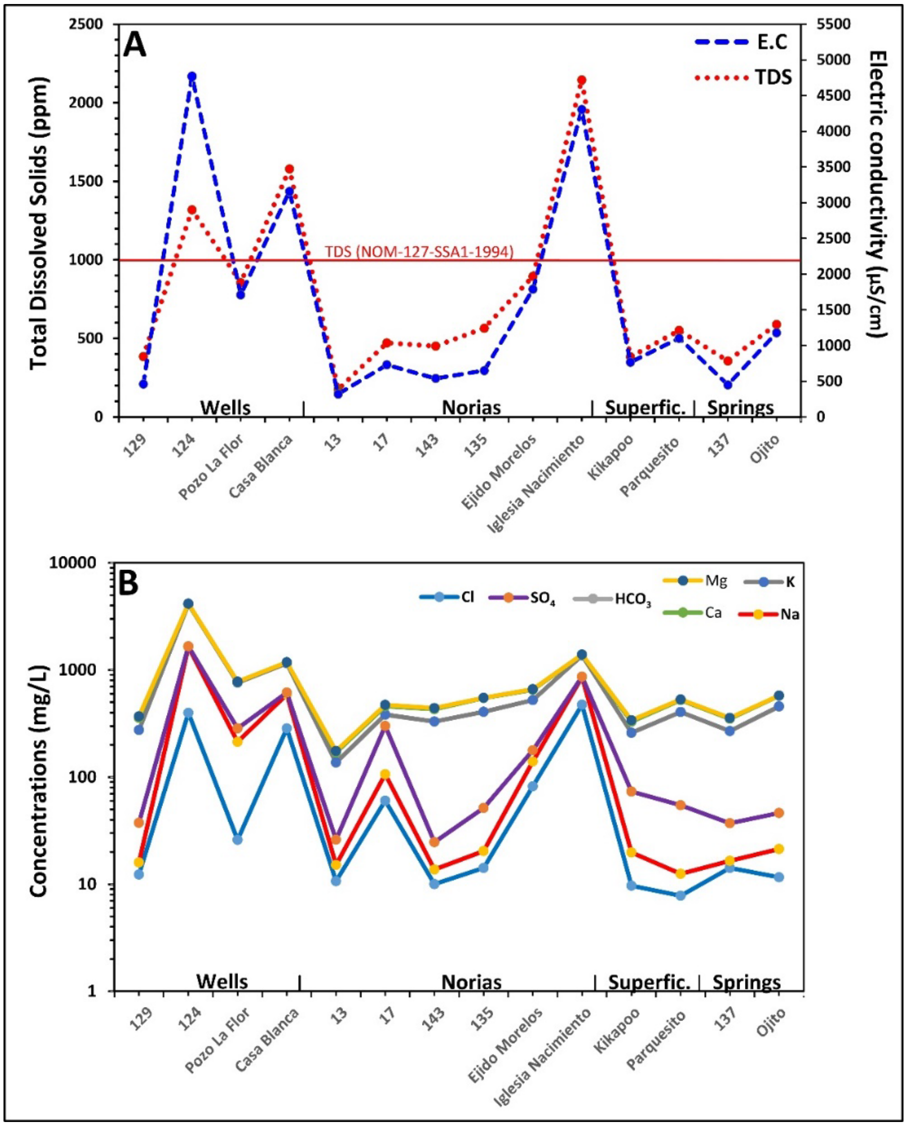

Figure 2. Comparative chemical analysis of water samples. A) Content of total dissolved solids and electric conductivity, and B) Concentrations of the major elements in the water. E.C.: Electric conductivity; TDS: Total dissolved solids.

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Concerning to the chemical composition of the water (Figure 2B), it is observed that the most enriched samples in the majority constituents are wells 124 and Casa Blanca, as well as the Iglesia Nacimiento noria. The highest Cl^- contents are reported as follows: Iglesia Nacimiento (2 146 mg/l), Pozo Casa Blanca (1579 mg/l), and Pozo 124 (1 319 mg/l), showing an enrichment of 8.5, 6.3, and 5.2 times in comparison with the maximum permissible limit established by the Official Mexican Standard NOM-127-SSA1-1994 (Cl^- = 250 mg/l). For the Na^+ element, enrichments of up to 6 times are reported in the sample from well 124 with respect to the aforementioned water quality standard. In general, the superficial bodies and the springs present low concentrations in most of the chemical constituents, which shows that they are not being affected by deep processes.

Isotopically, the waters under analysis vary between -3.29 ‰ to -7.36 ‰ for the isotope $\delta^{18}O$, while values between -23.02 ‰ to -48.24 ‰ are reported for deuterium (Table 2). In general, it is observed that the surface waters (e.g., Kikapoo and Parquecito) present the most depleted values (i.e., lighter) in both isotopes in relation to the rest of the water bodies, while the sample more enriched (i.e., heavier) is the Noria ($\delta^{18}O$ = -3.29 ‰). The Pozo Aparicio is not in the study area, however, due to its location in the Sierra de Santa Rosa it can help understand the relationship of this mountain range with the hydrology and recharge processes that occur in the region.

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Table 2. Isotopic composition of oxygen and deuterium of the waters under study (MASL: Meters above mean sea level).

Samples	Description	$\delta^{18}O$ (‰)	δD (‰)	altitude (msnm)
Kikapoo	River	-7.36	-48.24	513
Parquecito	River	-6.66	-43.49	455
Ejido Morelos	Noria	-6.09	-38.83	492
Iglesia Nacimiento	Noria	-4.79	-31.73	489
Noria	Noria	-3.29	-23.09	448
Pozo La Flor	Well	-6.24	-39.83	513
Pozo Casa Blanca	Well	-5.33	-34.87	605
Pozo Aparicio	Well	-7.04	-44.62	605
Ojito	Spring	-5.02	-32.85	564
Divi	Spring	-5.38	-34.21	570
Meteoric water Múzquiz	Meteoric water	-7.23	-47.8	500

Source: The composition of the ocean was taken from Craig (1961).

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Discussion

Hydrogeochemical and isotopic processes that control chemical variability in water bodies

Figure 3 presents the regional hydrochemical map based on the different Stiff diagrams (Stiff, 1951), which offer an overview of the different types of water and their spatial location.

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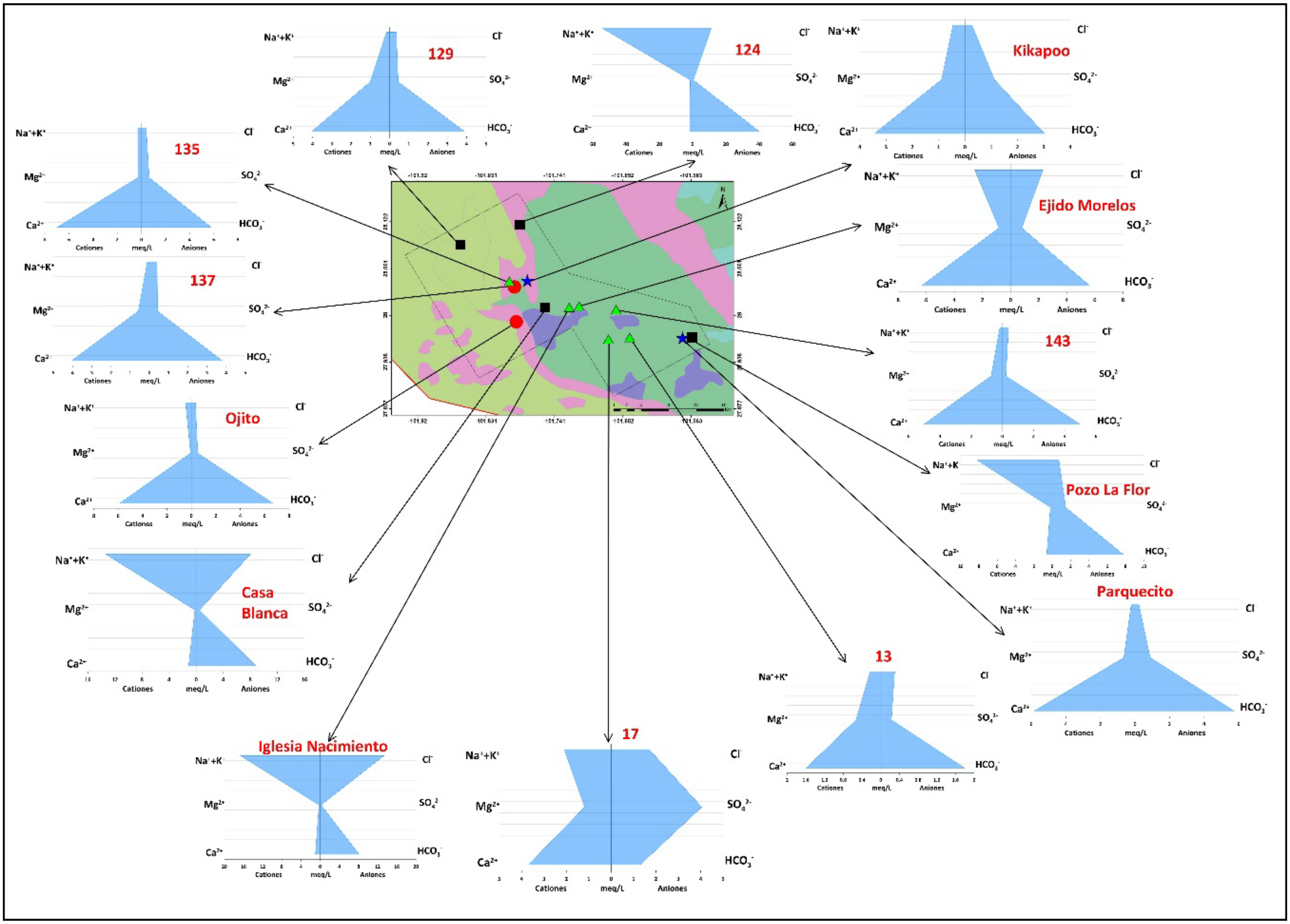

Figure 3. Map of the chemistry of the waters of the study area from the different Stiff graphs. On the map: black cubes-wells, green triangles-norias, red circles-springs, and blue stars-rivers and streams.

As indicated in the figure, nine water samples are bicarbonate-calcium and/or magnesium type, three samples bicarbonate-sodium type,

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1 sample of the chlorinated-sodium and/or potassium type and 1 sample was classified as Calcium Sulphated and/or magnesium. In the case of the Ejido Morelos noria, the pattern of the Stiff diagram, although mostly of the calcium bicarbonate type, has increases in chlorides and sodium, which suggests processes of increased salinity. In general, despite the study area only occupying a total area of 521 km^2, a great chemical variability is observed in the different water bodies, resulting from various geohydrological and environmental processes. In this sense, the Piper diagram (Piper, 1944) modified by Kelly (2006) offers a more complete interpretation.

Kelly (2006) defined the main hydrochemical processes responsible for chemical variability in water bodies, providing a different approach to Piper's (Piper, 1944) rhombus in which he established 7 new fields (Figure 4). The figure shows that the waters classified as bicarbonate-calcium (e.g., samples 129, 135, 137; Ojito, 13, Parquecito, 143 and Kikapoo) are located in the freshwater field, so from the point of view of water quality, are optimal for diverse activities. The composition of these waters comes from the dissolution of minerals present in carbonate rocks rich in Ca^{2+}, Mg^{2+} and, HCO_3^- (Choi *et al*., 2014), from La Peña and Aurora formations present in the Sierra de Santa Rosa. However, it is observed that the sample of the Ejido Morelos noria moves away from the vertex with a tendency toward the field defined by Kelly (2006) as "Light conservative mixture" due to saline intrusion observed in Figure 4.

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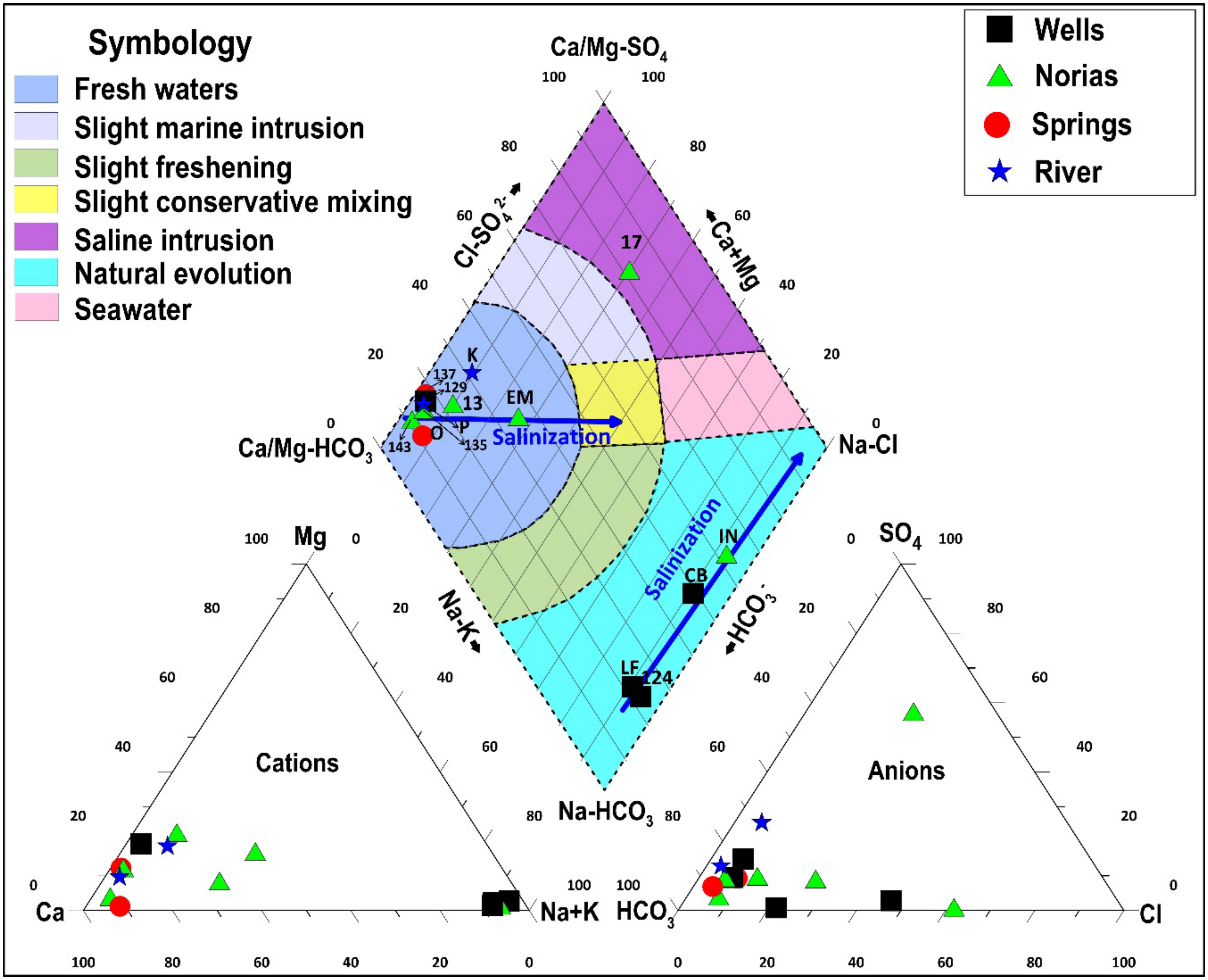

Figure 4. Kelly diagram (Kelly, 2006) showing the main hydrochemical processes that control the chemistry of the waters.

Bicarbonate-sodium type waters are reported for wells La Flor, 124 and Casa Blanca (Figure 4). The increase in sodium concentration in these waters seems to be related to water-rock interaction processes where

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there is high ion exchange, probably between clays, an important source of sodium supply. These sequences could be present in depth, interacting with the flows of the aquifer. In the case of the Casa Blanca well and the Iglesia Nacimiento noria, a tendency towards the vertex of sodium chloride waters is observed, which indicates salinization. In this case, and because they are wells and norias, the dissolution of existing salts in the geological environment, probably of an evaporite nature, very common in the north of Coahuila (Padilla-y-Sánchez, 2007; González-Sánchez, Puente-Solís, González-Partida, & Camprubí, 2007; Eguiluz-de-Antuñano, 2001), could justify said process and, therefore, responsible of the increase in the concentrations of Cl^-, Na^+, TDS and the electrical conductivity previously discussed. Additionally, surface processes such as evaporation or infiltration of highly evaporated water and solutions, could also induce salts into the environment, since these are semi-arid areas where evaporation is dominant over precipitation (Batista-Cruz *et al*., 2022).

A single sample (e.g., Noria 17) is classified as Sulphate-calcium and/or magnesium type water and is located in the saline intrusion field (Kelly, 2006). This sample may owe their composition to different processes, among which a) washing of land formed in the marine environment, b) oxidation of sulfides widely distributed in igneous and sedimentary rocks, and c) decomposition of organic compounds. In the study area, processes a) and b) are feasible because, as stated above, the geology of the state presents evaporite sequences of the old restricted marine conditions that prevailed in the Sabinas Basin from the Triassic to the Upper Cretaceous (Eguiluz-de-Antuñano, 2001), to which the study

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area belongs. The existence of geological sequences with rocks and soluble minerals such as gypsum, anhydrite, halite and magnesium, and potassium salts is widely documented throughout the region, representing a problem for water quality in several municipalities (Batista-Cruz *et al*., 2022). However, the oxidation of sulfides is also a non-negligible way of contributing sulfate to the water. In this sense, the Sierra de Santa Rosa and a large part of the geology of Coahuila have a wide distribution of metallic and submetallic mineral deposits with important galena and iron mines in operation, so both processes seem to be feasible as sulfate contributors to the waters of the study area.

Based on the diagram of Mifflin (1988) (Figure 5), the different types of groundwater flows were identified based on the concentration of Na^+, K^+, Cl^- and SO_4^{2-} ions. As can be seen in the graph, the samples under study are located in the three flows identified by the author (i.e., local, intermediate, and regional).

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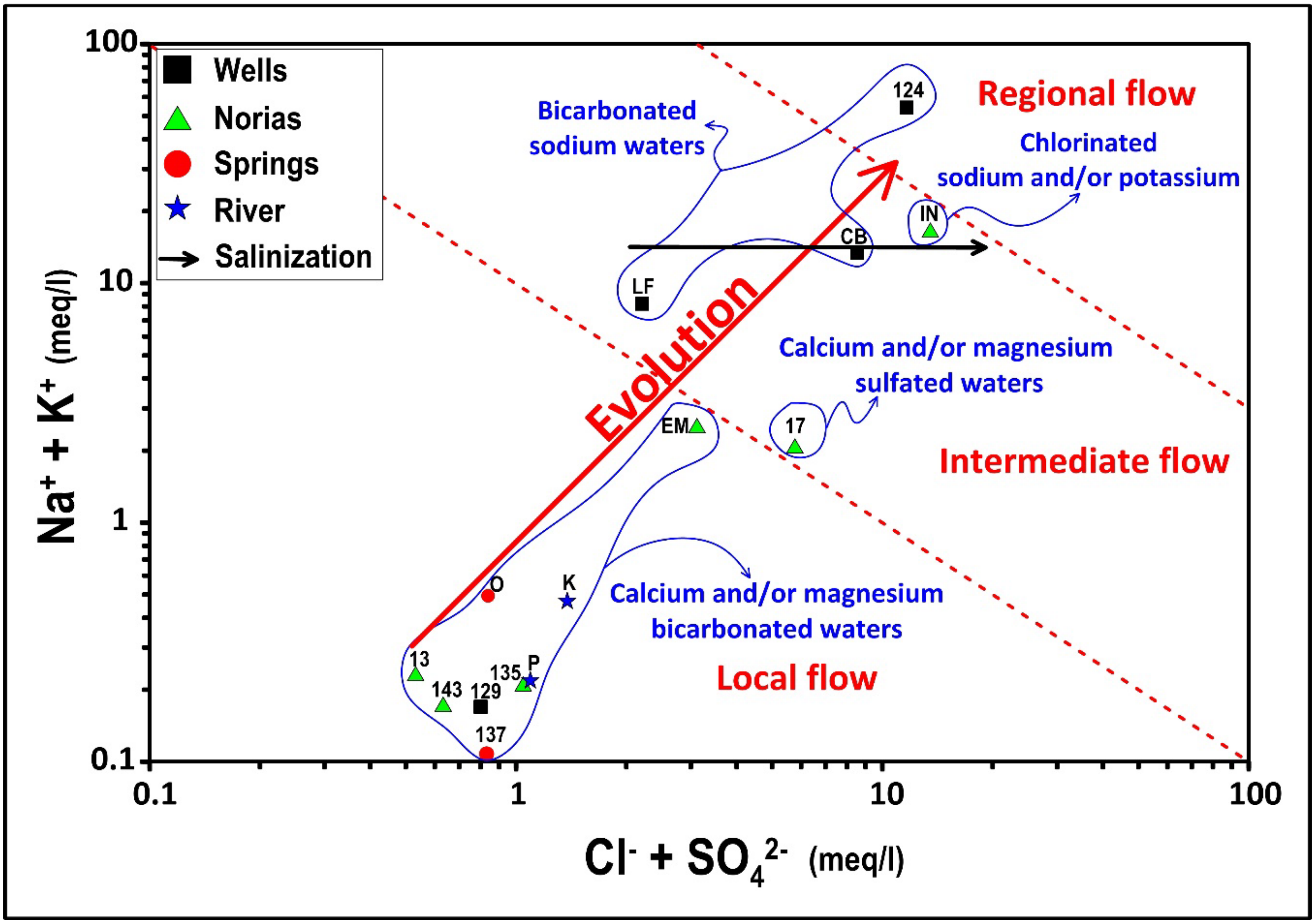

Figure 5. Binary diagram of Mifflin (1988) indicating the origin of the local, intermediate and regional flows. The figure also details the different types of water identified in the Stiff and Piper diagrams for a better understanding.

The waters belonging to the local flow (e.g., springs 137 and Ojito, well 129, norias 13, 143, 135 and the surface water samples Kikapoo and Parquecito) correspond to little evolved waters of recent infiltration, which coincide with a bicarbonate-chemical nature, and with the lowest values

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of the Na^++K^+ and Cl^-+SO_4^{2-} ratios. The Ejido Morelos sample, which had previously been identified in the Stiff and Piper diagram as having some salinity due to saline intrusion, corroborates its trend towards more mineralized waters located in the intermediate flows (Figure 5). The water samples from the La Flor and Casa Blanca wells, as well as the Norias 17 and Iglesia Nacimiento, are also located in this field. These waters present a greater hydrogeochemical evolution due to a longer residence time in the geological environment and the depth at which they infiltrated. In fact, waters from Iglesia Nacimiento are evolving towards the presence of regional flows. In general, all these water bodies have higher chemical concentrations as shown by the high values of Na^++K^+ and Cl^-+SO_4^{2-} compared to the rest of the samples (Figure 5). According to Chapelle (2000), intermediate flow systems generally correspond to confined aquifers of moderate depth (< 300 meters) and differ from local flow systems mainly in the extension of the connection with the surface and the saturated zone.

Finally, the waters of well 124, which are the most enriched in Na^+ and HCO_3^- (see also Table 1 and Figure 4), are chemically located within the regional flow, so they correspond to a solute deposition zone, where the water is it saturates the mineralization with product ions along the path traveled by the subsoil. Thus, they are considered the oldest, most mature, and evolved waters (Mifflin, 1988).

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Isotopic composition of waters

In hydrochemical studies, the use of stable isotopes (i.e., atoms of the same element with the same atomic number and different mass number) has positively influenced the interpretation of the processes that are part of the hydrological cycle and, therefore, allow clarifying the origin of water and the fractionation processes that affect its composition (Clark & Fritz, 1997; Mook, 2001). In order to integrate the isotopic information with the hydrogeochemical composition, this section presents the water's origin and a condensed conceptual model for the study area. Unfortunately, isotopic data is unavailable for all the samples, so the approach must be taken as general.

The oxygen *versus* deuterium isotopic graph in Figure 6 shows the different fractionation processes present in the water bodies under study. Surface samples from Kikapoo ($\delta^{18}O$ -7.36 ‰; δD -48.24 ‰) and Parquecito ($\delta^{18}O$ -6.66 ‰; δD -43.49 ‰), the Aparicio ($\delta^{18}O$ -7.04 ‰; δD -44.62 ‰) and La Flor ($\delta^{18}O$ - 6.24 ‰; δD -39.83 ‰) as well as the Ejido Morelos noria ($\delta^{18}O$ -6.09 ‰; δD-38.83 ‰) have a meteoric origin, coming from the local rainfall discharged in the Sierra de Santa Rosa ($\delta^{18}O$ -7.23 ‰; δD - 47.8 ‰) as shown by its location on the Global Meteoric Water Line from (Craig, 1961). The slight isotopic changes towards heavier isotopic values observed in the Pozo La Flor and the Ejido Morelos noria in relation to meteoric water, are probably due to differences in the altitude, water-rock interaction processes or ion exchange (Giggenbach, 1991).

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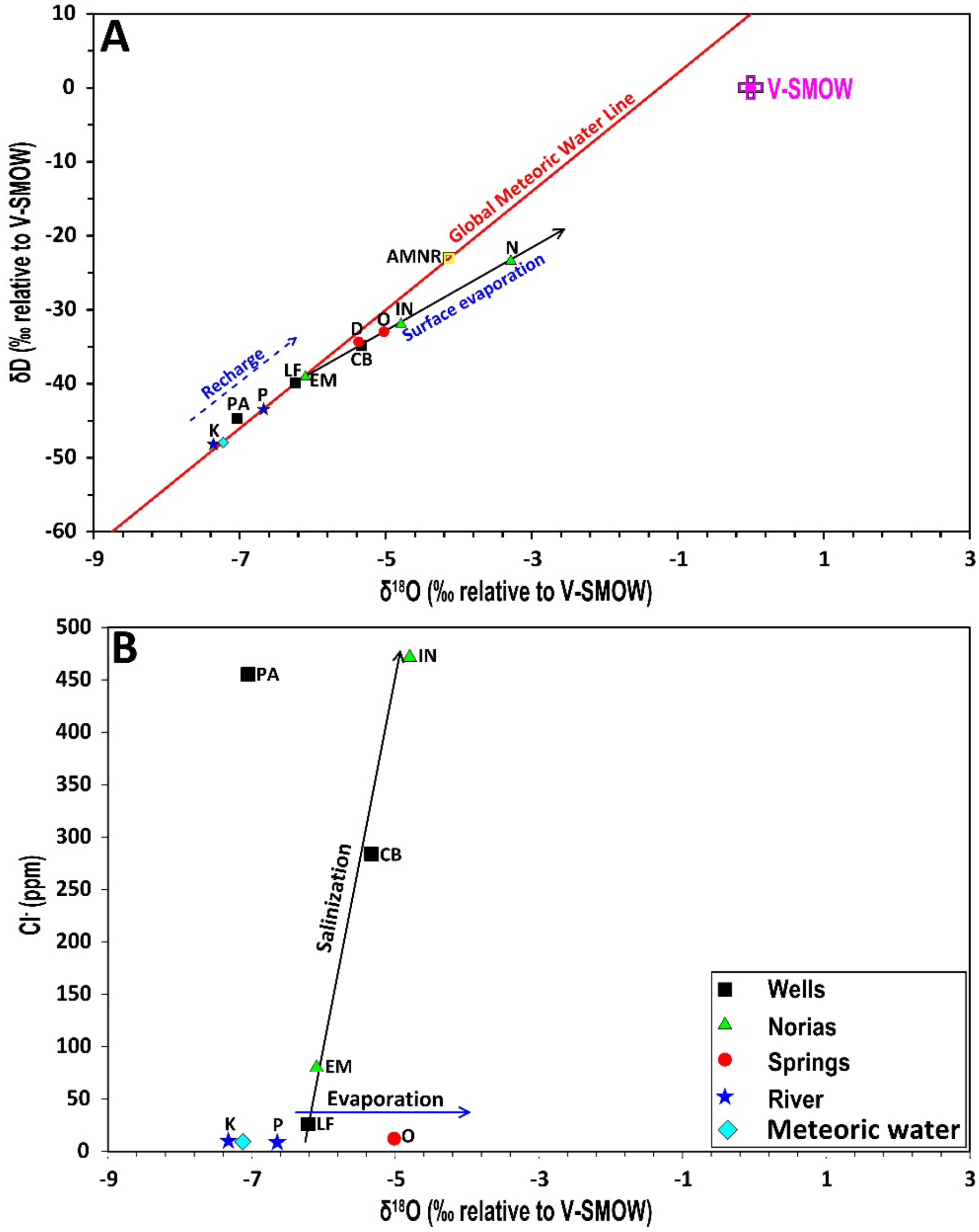

Figure 6. A) Oxygen *versus* deuterium isotopic plot where the Global Meteoric Water Line and the isotopic composition of the ocean (VSMOW) have been plotted, both taken from Craig (1961); B) Graph $\delta^{18}O$ *versus* Chlorides. AMNR-Nueva Rosita meteoric water.

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The water bodies with isotopic signatures heavier than local meteoric waters are Divi ($\delta^{18}O$ -5.38 ‰; δD -34.21 ‰), Casa Blanca well ($\delta^{18}O$ -5.33 ‰; δD -34.87 ‰), Ojito ($\delta^{18}O$ -5.02 ‰; δD –32.85 ‰, Iglesia Nacimiento ($\delta^{18}O$ -4.79 ‰; δD -31.73 ‰) and Noria ($\delta^{18}O$ -3.29 ‰; δD -23.09 ‰). Initially, these waters must come from local rainfall but have been fractionated by surface evaporation processes and/or dissolution of existing minerals in the crust. Due to the intense surface evaporation, the limited rainfall in the region, and the limited depth of these perforations (they do not exceed 50 meters), these processes seem to be feasible. Figure 7 and Figure 8 show the geological, hydrological and environmental particularities that seem to control the chemistry of the waters in the study area. From the geological point of view, 12 formations outcrop with ages ranging from the Lower Cretaceous to the Neogene and two Quaternary innominates lithologies of the Holocene composed of alluvial sedimentary deposits (SGM, 2003). The origin of the salinity and the enrichment in some ions, such as sulfate, could be related to the conditions of deposition and the sedimentary environments that gave rise to these geological formations. As shown in Figure 7, in the Sierra de Santa Rosa, the Santa Ana Anticline and the Loma Gorda Anticline, the formations have a sedimentary origin, with a predominance of limestone and shales deposited in shallow (formacions Cupido and Aurora) and deep (La Peña formation) marine environments. Coastal estuarine type sabkha is typical from the La Mula formation (Figure 7). At the base of the mountains and plateaus, the Buda, Eagle Ford, Austin, Upson, San Miguel and Olmos formations outcrop, whose petrological composition is mainly shale, siltstone and clayey, with the

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presence of limestone. Sedimentation environments vary from neritic to transitional deltaic or prodeltaic type.

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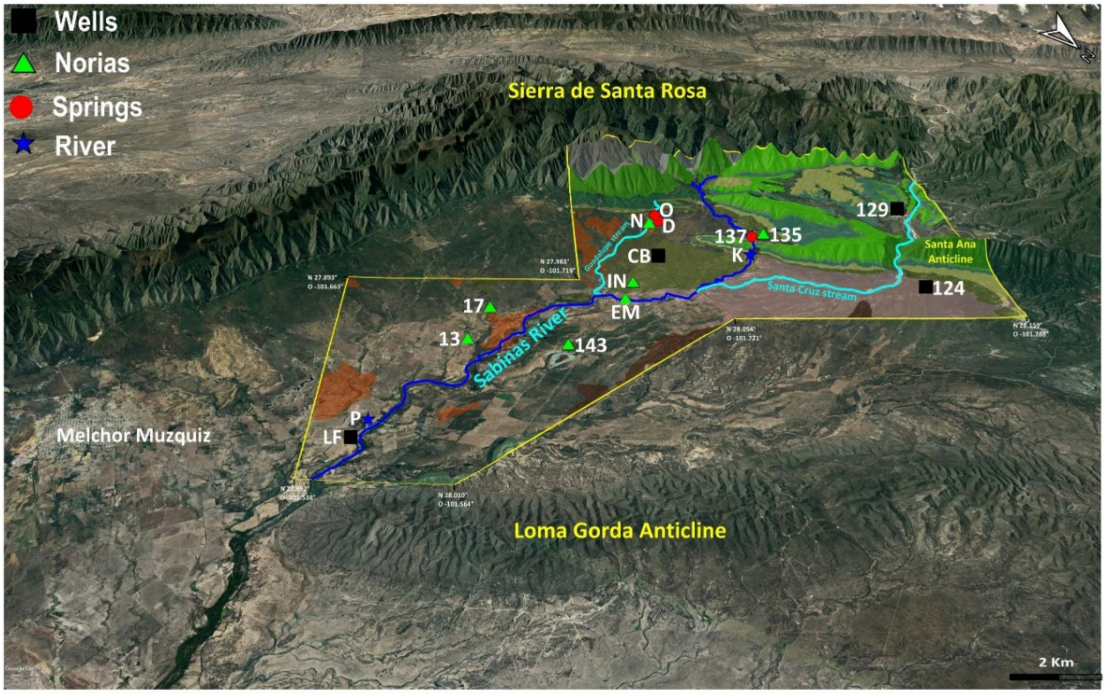

Geology				Sedimentary environments
Holocene	Qhoal	Alluvial deposits		
Holocene	QpthoCgp	Polygenetic conglomerate		Polygenetic conglomerate (continental fluvial)
Neogene	TplCgo	Fm Sabinas		Fm Sabinas (continental fluvial)
Upper Cretaceous	KmLu-Ar	Fm Olmos		Fm Olmos (Shallow marine waters and deltaic)
	KcAr-Lu	Fm San Miguel		Fm San Miguel (deltaic)
	KcLu-Lm	Fm Upson		Fm Upson (shallow plataform and prodelta)
	KcossCz-Lu	Fm Austin		Fm Austin (neritic with terrigenous contribution)
	KcetLu-Cz	Fm Eagle Ford		Fm Eagle Ford (shallow marine waters)
	KceCz	Fm Buda	KaceCz-Lu Washita Group	Fm Buda (neritic)
Lower Cretaceous	KceLu-Cz	Fm Del Río		Fm Del Río (neritic)
	KaCz-Do	Fm Aurora		Fm Aurora (shallow plataform)
	KapCz-Lu	Fm La Peña		Fm La Peña (deep water in bathyal zones)
	KapCz	Fm Cupido		Fm Cupido (shallow plataform)
	KhbLu-Cz	Fm La Mula		Fm La Mula (marine to estuarine sabkha)

Figure 7. Geological map of the study area made with the geological base 1:250,000 of the SGM (2003). In the legend of the map, the existing formations in the area are presented, and the sedimentation environments are synthesized according to the Mexican stratigraphic lexicon taken from SGM (2003). The alluvial deposits lithology (Qhoal) is represented without background color on the map.

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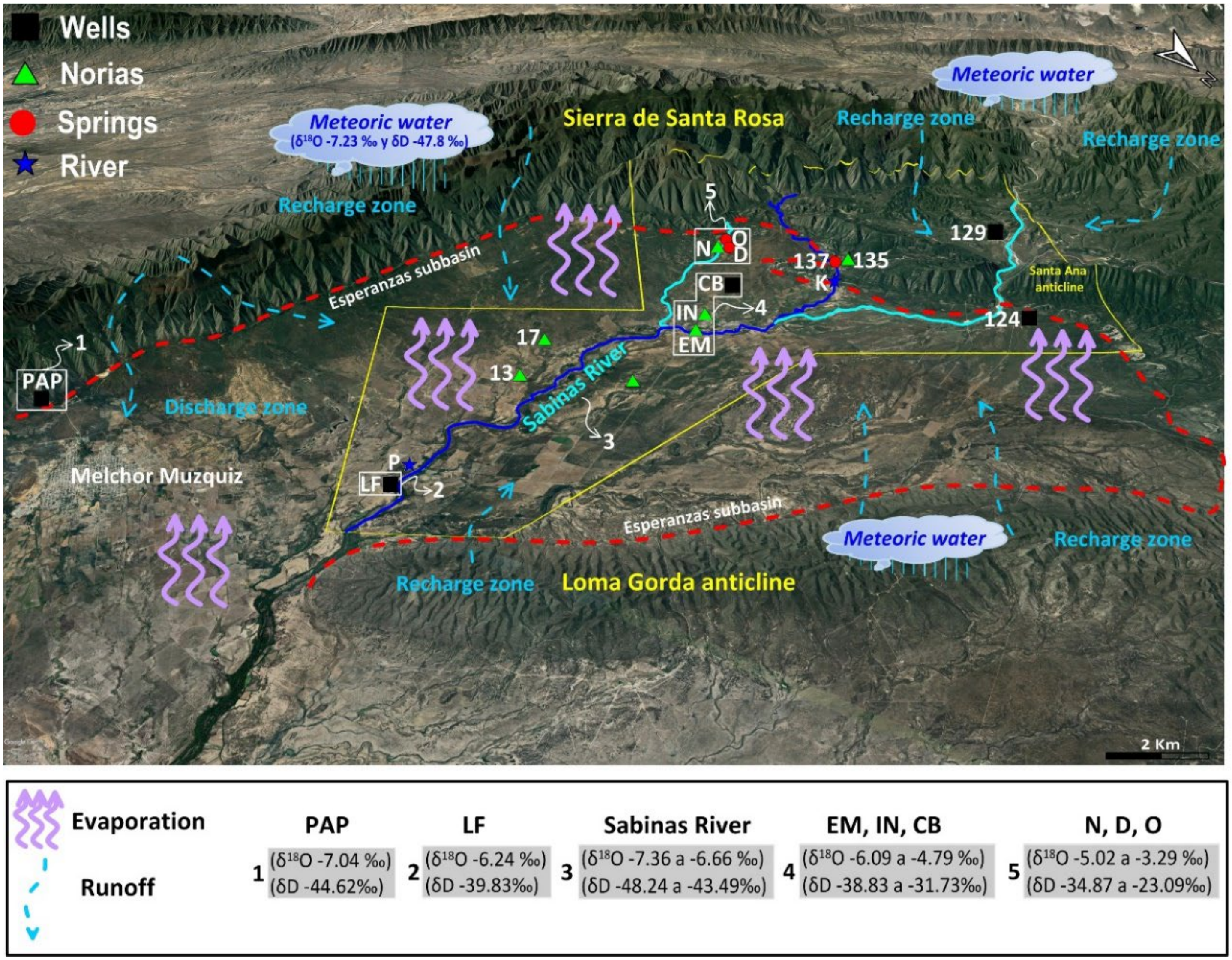

Figure 8. Conceptual geohydrological and isotopic model of the study area representing the main hydrological and environmental processes that control the chemistry of the waters. The letters in the lower legend are referred to in the text.

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Numerous works have referred to the presence of salts and minerals such as gypsum in some of these formations in various parts of the Coahuila state. According to Santamaría *et al*. (1991) in the La Mula formation, for example, there are horizons of soluble minerals such as anhydrite and gypsum. This formation outcrops in our study area, which suggests that the surface and underground flows of the Sierra de Santa Rosa could leach said minerals and contribute Ca^{2+} and SO_4^{2-} to the waters, as occurs in the sample noria 17. This process has been previously reported in some mountain ranges that surround the Cuatrociénegas area (Mamer & Newton, 2017) and in the Sierra Menchaca (Czaja, Estrada-Rodríguez, & Flores-Olvera, 2014).

In the case of evaporites, the existence of formations formed in deltaic, estuarine, and semi-restricted shallow marine environments (Figure 7) could contain minerals such as halite (NaCl) in their strata or sequences. However, although it appears to be feasible in theory, at least in the mountains surrounding the study area, there are no previous reports or field observations on the presence of this mineral or other minerals that contribute Cl^- and Na^+ to the water, therefore which seems to be superficial processes. No salts are reported in the alluvial valleys formed by the Sabinas formation, the polygenetic conglomerates, and the alluvial deposits.

The surface processes of an environmental nature are presented in the model of Figure 8.

The model begins with the fall of meteoric waters on the mountains surrounding the Esperanzas subbasin, where the study area is located (Figure 8). The recharge-discharge processes allow the valleys and plains

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to obtain significant volumes of water and thus, the initial isotopic signatures of the meteoric type ($\delta^{18}O$ -7.23 ‰; δD -47.8 ‰) are modified towards slightly heavier values in both isotopes. This is evident in the wells PAP ($\delta^{18}O$ –7.04 ‰; δD –44.62 ‰) and LF ($\delta^{18}O$ –6.24 ‰; δD -39.83 ‰). These signatures are consistent with infiltration processes in the carbonate sequences and ion exchange. For its part, the isotopic composition of the Sabinas River ($\delta^{18}O$ –7.36 to -6.66 ‰; δD -48.24 to –43.49 ‰) taken from the Parquecito and Kikapoo samples, is in the range of the composition of meteoric water, which confirms their close relationship as part of the hydrological cycle of the area. The rest of the water bodies with available isotopic data undergo evaporation processes, which fractionate both isotopes towards heavier values (Horita, 2005). This is observed in the Ejido Morelos, Iglesia Nacimiento and Casa Blanca samples (Point 2 in Figure 8) as well as in the Noria, Ojito and Divi samples (Point 3 in Figure 8). In summary, it can be shown that the processes of leaching and dissolution of mineral rocks, surface evaporation and solute transport increase the contents of SO_4^{2-}, Cl^-, Na^+ and other cations and TDS and electrical conductivity in all samples under study.

The data presented here show the hydrochemical complexity of water resources in semiarid subbasins, which indicates that geohydrological studies require a deep chemical and isotopic analysis to clarify the water's origin and their chemistry. The study also highlights the synergy that exists between hydrochemistry and isotopy in the identification of phenomena that affect water bodies and, therefore,

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serves as a case study for future work of this type in the State of Coahuila or outside of it.

Conclusions

Based on the data and arguments presented in this work, the following is concluded:

1. In the study area, different types of water were identified that respond to various geohydrological and environmental processes, for example:
   a) Bicarbonate-calcium type waters are identified for sites 129, 143, 13, 137, 135, Ojito, Parquecito and Kikapoo, whose composition reflects the dissolution of carbonates from the geological formations present in the Sierra de Santa Rosa and surrounding mountains.
   b) Sulfate-calcium and/or magnesium type waters are reported for noria 17 whose sulfate enrichment suggests saline intrusion processes or dissolution of evaporite rocks such as gypsum. Said mineral could be present in the La Mula formation that outcrops in the Sierra de Santa Rosa.
   c) Salinization processes are identified in wells LF, 124 and CB and wells IN, and EM. The increase in salts seems to be related to surface evaporation and subsequent infiltration of salts into the aquifer. However, it is not ruled out that the deltaic, estuarine or sabkha-type formations present in the area could contain soluble minerals such as halite with incidence in the aquifer.

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2. The Mifflin diagram showed the existence of different flows, from local to regional. The local flows correspond to calcium bicarbonated waters, the intermediate flows are those waters classified as calcium sulfate, sodium chloride and sodium bicarbonate, while the water from well 124 is considered within the regional flow since it is the most enriched sample in sodium and corresponds to a zone of solute deposition, where the chemical composition tends to increase its mineralization until it becomes saturated in the different ions along the path traveled through the subsoil.
3. From the isotopic point of view, a meteoric origin is proposed for most of the water bodies under study, whose recharge zone comes from the Sierra de Santa Rosa and the mountains that border the sub-account. The rest of the waters undergo surface evaporation and/or dissolution of salts, generating isotopic fractionation and heavier isotopic signatures.
4. The geological characteristics, the sedimentation environments and environmental processes that occur in subbasins of semiarid climates are determining factors in the variable chemistry observed in the waters, and in the deterioration of the quality of the resource.

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