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ORIGINAL ARTICLE

Identification of salinization processes in coastal aquifers using a fuzzy logic and data mining based methodology: study case in a Mediterranian coastal aquifer (Spain)

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Abstract

In coastal aquifers, the seawater intrusion can mask the effects of salinity regional groundwater flows, connate waters mobilization or contaminant process. Therefore, to discriminate between all the processes that have taken place in the coastal aquifer, is a complex task. Normally, traditional hydrogeochemical methods (e.g., Piper and Durov) together with statistical multivariate techniques (e.g., cluster and factorial analysis) and other methods (e.g., ionic deltas and isotopic studies) have been used to understand the hydrogeochemistry of aquifers and to confirm previous hypothesis. This paper presents a characterization of the salinization process in coastal aquifers, by means a fuzzy logic and data mining based methodology, which has not been used before for this purpose in a coastal aquifer. The proposed fuzzy methodology is based on the use of the data mining computer tool Predictive Fuzzy Rules Generator (PreFuRGe). The results have been obtained by processing groundwater samples analyses with PreFuRGe. The parameters used for the experimentation have been: temperature, electric conductivity, redox potential, total dissolved solids, silicon dioxide, oxidability, major ions (chloride, sulphate, bicarbonate, nitrate, calcium, magnesium, sodium and potassium), and minor ions (arsenic, bromide, lithium, boron, strontium, chromium and fluoride). The application of this method has made it possible to differentiate several overlapping hydrogeochemical processes, such as seawater intrusion, the entry of high salinity regional groundwater flows with high concentrations of strontium, magnesium, lithium and sulphates, and the effect of contamination from agricultural activities, with the presence of nitrates. The qualitative obtained results in this paper have been compared to previous research carried out in the same coastal aquifer, and it is proved that the used fuzzy methodology is a powerful tool for discriminating between overlapping geogenic and anthropogenic processes in coastal aquifers.

Keywords Seawater intrusion · Contaminant processes · Fuzzy logic · Data mining · Hydrogeochemical analysis · Mediterranean coastal aquifer

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Introduction

Coastal aquifer hydrogeochemical processes can be highly complex. It can be very difficult to discriminate between overlapping geogenic and contaminant processes, due to the effects of the dominant process, as seawater intrusion (Daniele et al. 2022). High salinity can be caused by the mobilization of connate waters (Lambrakis and Marinos 2003; Stoecker et al. 2013), high salinity regional groundwater flows, mixing of water from different origins, and the effects of contamination from human activities (Custodio 2010; Gassama et al. 2011; Mondal et al. 2011; Abu-Salem et al. 2022), especially irrigation for agriculture. Determining the causes of high salinity requires the application of many different research methods (García-Menéndez et al. 2016).

Traditional hydrogeochemical methods based on the creation of specialized graphs, which can be considered as an approach to cluster analysis (e.g. Piper, and Durov) and the study of ionic relationships (e.g. binary plots and hydrogeochemical indexes) are often adequate. Normally, these methods are capable of discriminating between salinity mechanisms, identifying water families, emphasizing trends, and characterizing the hydrogeochemical processes involved in water–rock interactions (Giménez and Morell 1997). The application of other methods, such as the analyses of hydrochemical facies, ionic deltas, and isotopic studies (Giménez and Morell 2008; Duque et al. 2011; Yang et al. 2011; Alfarrah and Walraevens 2018; Torres-Martinez et al. 2021), allows a deeper understanding of the spatial distribution of the water types and the evolution of the processes involved.

Frequently, hydrogeochemical analyses employ statistical multivariate techniques as cluster analysis, and factorial analysis. These analyses are often used to confirm previous hypotheses. In addition, these techniques allow ionic associations to be identified in very complex situations, or when there are large numbers of variables (minor and trace ions). These associations might not be detected using more traditional methods (Morell et al. 1996).

Other tools can also be used, such as those based on the use of tracers (Boumaiza et al. 2025; Giménez-Forcada and Vega-Alegre 2015a, b), isotopes (Kaown et al. 2021), in mixing models, which also use isotopes such as the Bayesian isotope mixing model (Boumiaza et al. 2025; Ju et al. 2022), as well as in machine learning models such as neural networks (Secci et al. 2022) or mathematical flow and transport models (Ncibi et al 2022). Each of these tools has a different degree of applicability and complexity, especially depending on the availability of data and the problem to be studied, so it is necessary to look for other tools that can facilitate this discrimination of the origin or origins of pollutants and that can also be used in combination with other tools such as those identified above. In general, the best results are obtained by combining diverse methods.

This paper presents a new approach to characterizing the salinization processes in coastal aquifers. The proposed fuzzy logic methodology is based on the data mining computer tool Predictive Fuzzy Rules Generator -PreFuRGe- (Aroba 2003), which has proved to be suitable for modelling the qualitative behaviour of complex systems (Grande et al. 2005, 2010; Davila et al. 2021). In the context of this research, the fuzzy logic has been used to address some problems such as changes in the spatial variability of hydraulic parameters (Das et al. 2016), identification of quality indexes of groundwater (Vadiati et al. 2016; Guo et al. 2020), distribution of hydrochemical facies (Güler and Thyne 2004; Güler et al. 2012; Agoubi et al. 2016; Rahbar et al. 2020), or prediction of a rock engineering classification system (Jalalifar et al. 2011).

This article proposes to check the possibility of using a fuzzy logic and data mining based methodology to differentiate between overlapping hydrogeochemical processes that take place in coastal aquifers, to propose its use as a complementary tool in hydrogeochemistry studies. The obtained results have been compared with previous works in the same study area, and it is checked the veracity and importance of the qualitative information generated by PreFuRGe from quantitative analyses data, for discriminating between overlapping processes in coastal aquifers.

Geological and hydrogeological setting

The Plana de Castellón is a Mediterranean coastal aquifer system (Plio-Quaternary detrital aquifer) located in eastern Spain, which has an area around 500 km^2 (Fig. 1). This aquifer is bordered by Desierto de las Palmas Mountains (Jurassic and Cretaceous units) to the north, by the Sierra de Espadán Mountains (Triassic units with the three characteristic facies: Buntsandstein, Muschelkalk, and Keuper (López-Gómez and Arche 1993) to the southwest, and by the Mediterranean Sea to the east. The study area is located in the south of the aquifer system (Fig. 1).

Materials of the study area consist of gravel, conglomerate, sand, and clays, with a maximum thickness of 120 m. In general, the aquifer system is detrital, shallow, and heterogeneous. Considered in detail, four geological layers can be identified (García-Menéndez et al. 2016): two detrital Plio-Quaternary layers separated by a 20 m thick, an impermeable clay layer, and a Mesozoic substratum (MS). Both have hydraulic connection. The detrital layers consist of an Upper Detrital Aquifer (UDA) and a Lower Detrital Aquifer (LDA). The UDA unit is 85 m thick, and

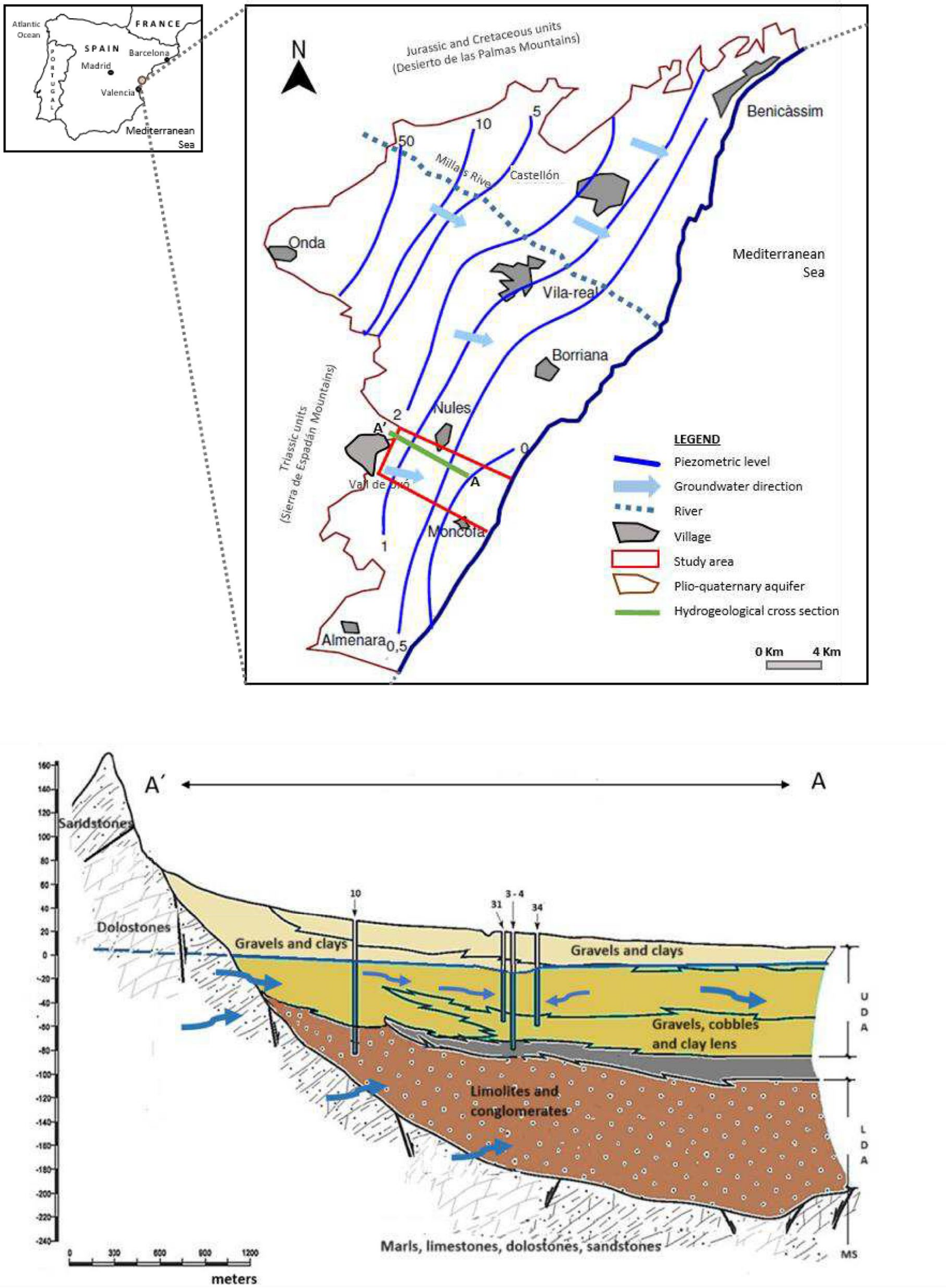

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Fig. 1 Study area location (**A**) and hydrogeological cross-section A–A' (**B**). UDA and LDA are Plio-Quaternary detrital aquifer. MS is Mesozoic Substratum. Both have hydraulic connection (adapted from García-Menéndez et al. 2016 and Renau-Llorens 2010) comprises gravels, sandstone, and clay layers. The lithology of the materials is heterogeneous, predominating the existence of carbonate pebbles (limestone) with a silt–clay matrix. The porosity is intergranular, hydraulic conductivity, and specific yield of 5%, 90 m/day, and 6–10 L/s/m, respectively. In contrast, the LDA (Lower Detrital Aquifer) unit is composed of limolites and clays with layers of sandstones and polygenic conglomerates. The thickness of the LDA is 50–100 m, hydraulic conductivity of 40 m/day, and increases towards the coast. The hydraulic conductivity of the UDA is higher than that of the LDA; thus, the LDA is of less hydrogeological importance (contributes less flow to the aquifer system). The Mesozoic Substratum (MS) underlies the above-mentioned formations. The MS comprises (Giménez-Forcada and Vega-Alegre 2015a, b): (i) Limestones, marls and dolostones (Muschelkalk facies). (ii) Materials accommodate frequent intercalations of red clays and evaporitic sulphate salts with orthoquartzitic sandstone and limolites (Buntsandstein facies). These facies have been characterized by presenting hydrothermal mineralizations and iron oxides and hydroxides. (iii) Marls with gypsum (Keuper facies). The substrate formed by Mesozoic materials (MS) presents horst and graben structures.

The permeable formations of MS (Muschelkalk dolostones and Buntsandstein sandstone) may be hydraulically connected to the Plio-Quaternary aquifer system (García-Menéndez et al. 2016; Morell et al. 1996) (Fig. 1B).

In general, the natural groundwater flow in the Plana de Castellón aquifer system is from inland to the coast (Fig. 1A). However, in the study area, the direction of groundwater flow has been changed because of intensive extraction (Fig. 1B). In the study period, detailed piezometry is shown in Figs. S1 and S2 of the supplementary information.

The extraction began a few decades ago (1980s) and intensive salinization process of groundwater (seawater intrusion) has been induced (Giménez 1994; Giménez and Morell 2008; Renau-Llorens 2010). The geometry of the freshwater-seawater contact is caused by the groundwater extraction. The pumping wells have flowrates between 30 and 100 L/s. The result has been an upconing formation with a diameter of 3000 m (surface that can be delimited based on hydrochemical data through an isovalues map). It is located between the villages of Nules, Moncófar, and Vall de Uxó. The results and conclusions of Garcia-Menendez et al. (2016) support the hydrogeochemical processes and dynamics of seawater intrusion and upconing.

The water budget is characterized by intensive extraction from pumped wells. The pumping exceeds the recharge to the system. Inputs to the system include lateral groundwater inflow from the boundary aquifers (MS and Triassic units of Sierra de Espadán, there is a hydraulic connection between these fractured aquifers and the detrital aquifer system), the infiltration of precipitation, unplanned recharge (which comes from agricultural irrigation), from groundwater or regenerated water from the Vall de Uxó wastewater treatment plant; and seawater intrusion. Outputs include discharge to the sea and pumping. The system is currently in a state of unbalance caused by intensive crop irrigation (with greater discharge than recharge). Morell et al. (2009) estimate for the southern sector, in which the study area is located, a lateral inflow of 19.8 hm^3/year, an infiltration of 12.4 hm^3/year, internal transfers of 0.9 hm^3/year, and inputs due to seawater intrusion of 6.0 hm^3/year, while discharge by pumping are 31.1 hm^3/year, to the sea 0.2 hm^3/year, by internal transfer 1.4 hm^3/year and through wetlands 6.5 hm^3/year.

The hydrogeochemical characteristics in the study area are the result of an overlap of different natural processes, such as the discharge of sulphate groundwater flows from the boundary aquifers (MS and Triassic units of Sierra de Espadán), and human processes that have led to seawater intrusion and agricultural and industrial pollution. Consequently, it is possible to distinguish two water types, depending on their salinity: chloride water from the sea and sulphate water from inland (Fig. 2). Chloride groundwater is characterized by high chloride, sodium, and bromide concentrations that are compatible with the mixing of freshwater of recent infiltration (rainwater infiltration and unplanned recharge, which comes from agricultural irrigation) and seawater (Giménez and Morell 2008, Morell et al. 2008; García-Menéndez et al. 2016, Almazán et al. 2024). Sulphate groundwater is characterized by magnesium, lithium, sulphate, and strontium concentrations that cannot be accounted for by seawater intrusion (Fidelibus et al. 1993; Giménez 1994; Giménez and Morell 2008, López Gutiérrez et al. 2010; Giménez-Forcada and Vega-Alegre 2015a, b). Water–rock interaction processes contribute to a substantial modification of the composition of the water, especially by inverse ionic exchange. The calcium concentration of the water at the expense of sodium by ionic exchange has been increased by water–rock interaction and by bicarbonate dissolution. The high nitrate concentrations are caused by crop activity (Renau-Llorens 2010), while some of the heavy metals originate from industrial activity and/or inland aquifers (Escrig et al. 1993, López Gutiérrez et al. 2010). Thus, a complex hydrogeochemical scheme is created.

The spatial distribution of the two water types is shown in Fig. 2A. The chloride waters occur in the west where intensive extraction is caused by pumping wells. This extraction causes significant upconing of saline water (García-Menéndez et al. 2016). The sulphate waters are located close to the coast, where local discharge from MS through horst-graben structures is well known (Fidelibus et al. 1993; Giménez-Forcada and Vega-Alegre 2015a, b) (Fig. 2B).

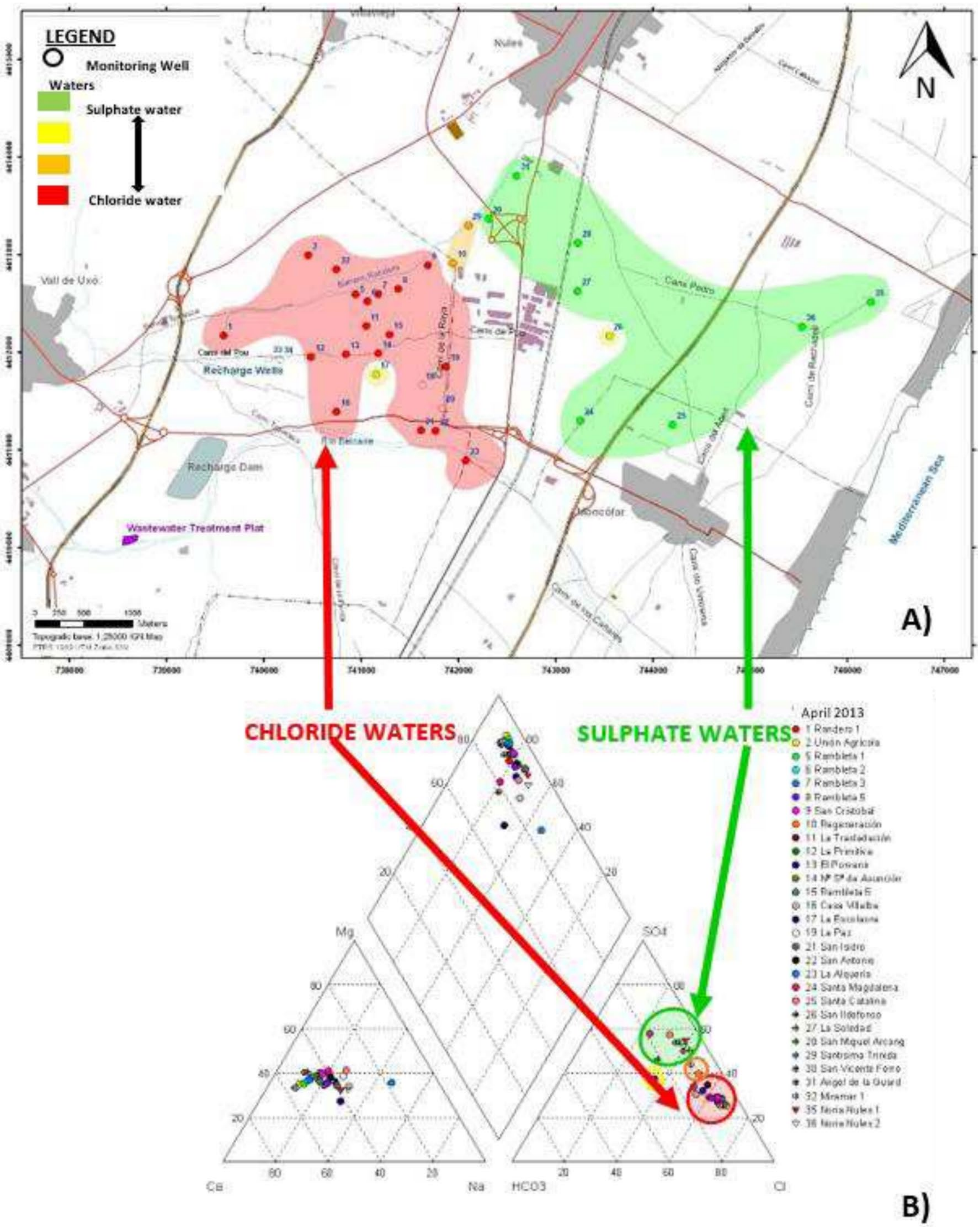

Fig. 2 **A** Monitoring network location and hydrochemical facies. **B** Piper diagram (April 2013). Sulphate facies (green), chloride facies (yellow), sulphate chloride facies -2 families- (orange and red) (adapted from Morell et al. 2013)

Materials and methods

Groundwater monitoring network and samples

The groundwater monitoring network consists of 37 wells distributed, across a surface of approximately 32 km^2, of which water samples were taken in 31 wells (Fig. 2A). Samples are from wells representing groundwater flowing through the UDA unit. Most of the wells are agricultural wells but some have been abandoned because of poor water quality (of the 37 wells inventoried, 16 are abandoned).

Two sampling campaigns (in April 2013 and April 2014) were carried out. In each campaign, samples were taken 5 m below the piezometric level using a Solinst 425 sampler. Field measurements of pH, temperature (T), and electric conductivity (EC) were taken using a Eutech Instruments PC650 portable probe. The redox potential (Eh) was measured in situ with a Crison pH meter 507. The field equipment was calibrated using pH solutions of 4, 7 and 10, electrical conductivity solutions of 1413 µS/cm, 5000 µS/cm and 12.88 mS/cm and Eh solution of 240 mV.

The water samples were filtered in situ with a 45 µm filter, stored in polythene bottles, acidified for the case of cation analyses, and sent to the Spanish Geological Survey (IGME) and the University of Jaume I (UJI) of Spain to be analyzed. The obtained data are shown in Table S1 and Table S2 of the Supplementary Information.

Chloride (Cl^-), sulphate (SO_4^{2-}), bicarbonate (HCO_3^-), nitrate (NO_3^-), calcium (Ca^{2+}), magnesium (Mg^{2+}), bromide (Br^-), silicon dioxide (SiO_2), total dissolved solids (TDS), and fluoride (F^-) were analyzed by a molecular absorption spectrophotometry-continuous flow analyzer (Alliance Integral Futura). Sodium (Na^+) and potassium (K^+) were determined by atomic emission spectrometry (Varian Spectra AA-220, model 330 FS). The analysis of arsenic (As), lithium (Li^+), boron (B), strontium (Sr^{2+}), chromium (Cr), and oxidability (Oxi) was performed using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent model 4500 with a Babington type nebulizer).

Variance of duplicate measurements was less than 10%; recoveries of checked and spiked samples were between 90 and 110%. The physicochemical results were validated by estimating the ionic balance error, which is the electroneutrality condition. The charge balance errors for the analyses were generally $\pm 5\%$, which is an acceptable error for the statistical treatment of a hydrochemical data set (Freeze and Cherry 1979).

The information obtained in the campaigns for both years (April 2013–April 2014) represents two different years with similar piezometric levels and hydrochemical characteristics (García-Menéndez 2018). To correct for deviations, the average values for the two campaigns were used (Table 1).

Data mining and fuzzy logic

Data Mining is used to extract meaningful (and sometimes unknown) information from databases. Computer tools based on this technology enable the systems characterization and the prediction of behaviors, based on the used stored data. The results obtained with data mining based tools can go further than the knowledge provided by the classical statistical techniques (Hand 1998; Glymour et al. 1996). From the expert point of view, it is very useful to be able to analyze the obtained results qualitatively, by using natural language. In this way, using Fuzzy Logic, the obtained results can be interpreted in the same way as a human would do (Furuhashi et al. 2001; Guillaume 2001; Nakajima 1996).

Fuzzy Logic (Zadeh 1965) operates using rules of reasoning very close to the human way of thinking and allow to characterize a variable without specifying a precise value (not possible with classical logic). In Classical Logic, an element is either a member or not of a set (binary membership). Conversely, Fuzzy Logic allows to associate each element with a set, with an accurate membership grade (using a membership function).

Fuzzy clustering

A clustering algorithm classifies a dataset in groups (clusters), where the elements in each group are more similar to each other than to those in other clusters (Kaufman et al. 2009). In classical clustering algorithms, each element of the dataset is assigned to a cluster, where the membership grade of each element can only take values 0 or 1, indicating null or whole membership to each cluster.

On the other hand, in fuzzy clustering algorithms (Hathaway and Bezdek 1993), the membership grade of each element can take real values between 0 and 1 (partial membership grade). Therefore, elements may belong to more than one group (cluster), so that the clustering capacity increases. A well-known fuzzy clustering algorithm is the so-called Fuzzy C-Means (FCM), it is based on the minimization of distances between two elements and the prototypes of cluster centers (Hathaway and Bezdek 1993; Fukuyama and Sugeno 1989; Hoppner and Klawonn 2003; Rahbar et al. 2020).

Predictive fuzzy rules generator (PreFuRGe) methodology

The first step, before data processing, is to determine the parameters whose behavior researchers want to analyze. In the fuzzy rules that will be obtained, these parameters will

Table 1 Average values of physicochemical parameters (April 2013–2014) used in the Fuzzy Logic analysis

ID	EC	T	pH	Eh	TDS	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}
1	2600	21.7	7.17	197	1932	122	9	294	134	576	333
2	2990	17.8	7.21	199	2500	113	3	449	160	820	366
5	2390	18.0	6.36	232	2274	113	3	390	160	676	400
6	2980	18.0	7.39	228	2351	120	3	378	171	660	390
7	2680	18.1	6.14	225	2419	134	4	390	180	720	420
8	2540	18.9	7.07	224	1774	129	5	266	113	544	283
9	3290	21.7	7.08	159	2726	168	4	371	221	760	483
10	2775	19.8	7.09	215	2225	127	3	334	156	452	527
11	2695	18.1	7.35	230	2052	113	3	327	146	544	320
12	2805	17.8	6.90	214	2153	135	5	323	149	544	351
13	2635	18.3	6.96	216	2008	159	5	271	137	520	365
14	2490	18.2	6.60	199	2293	200	6	310	170	690	410
15	3105	17.8	7.12	242	2373	158	5	357	160	660	417
16	2665	20.0	7.03	214	1994	187	7	262	127	648	350
17	2122	19.6	7.29	155	1519	134	6	207	88	628	266
19	3640	18.6	7.26	189	2974	314	4	348	216	852	536
21	2870	19.5	6.17	217	2720	275	7	360	190	890	460
22	2965	17.6	7.47	259	2429	194	3	315	170	604	518
23	2440	18.2	7.17	222	1852	319	4	108	130	470	410
24	1592	18.6	7.07	263	2459	89.5	8	156	88	126	438
25	3025	18.6	7.15	215	2503	257	6	274	169	468	754
26	2155	17.8	7.17	152	1933	130	9	235	105	326	500
27	3185	19.0	6.89	230	2820	187	7	378	198	540	801
28	2775	18.4	7.01	227	2277	127	3	340	151	344	655
29	2710	17.9	7.17	218	2179	105	2	303	170	410	549
30	2960	19.1	7.07	225	2199	180	5	322	160	692	528
31	2240	18.9	6.76	231	2227	85	3	390	140	340	680
32	2360	18.8	6.80	194	2313	96	7	205	75	680	175
35	3580	17.3	7.01	206	3319	362	7	422	185	570	1225
36	3010	16.4	7.34	199	2801	323	7	319	160	460	910
37	2820	18.8	7.76	160	2081	76	6	146	74	528	186

ID	HCO_3^-	NO_3^-	SiO_2	F^-	Br^-	Oxi	Li^+	B	Cr	As	Sr^{2+}
1	187	320	13.9	0.20	1.10	1.3	15.1	99.45	0.48	0.24	1291
2	147	435	11.2	0.17	1.62	2.7	14.5	69.30	0.43	0.40	1214
5	101	420	11.1	0.11	1.30	5.5	< Q.L	46.60	0.49	0.25	899
6	161	445	12.6	0.18	1.35	1.7	14.9	74.95	0.99	0.36	1398
7	119	440	12.5	0.17	1.30	2.1	< Q.L	62.70	1.31	0.23	1189
8	167	295	12.9	0.20	1.09	1.4	114.0	91.30	0.38	0.19	1724
9	227	470	15.5	0.22	1.59	1.9	36.8	122.50	7.77	0.15	1341
10	204	390	14.6	0.19	0.57	1.1	49.1	137.00	6.80	0.16	1912
11	161	375	14.0	0.15	1.17	1.6	11.6	49.75	0.96	0.25	945
12	189	335	15.0	0.16	1.31	1.7	8.6	48.53	0.62	0.21	1233
13	224	315	15.5	0.19	1.03	1.4	8.4	70.65	2.01	0.20	995
14	144	370	12.9	0.13	1.27	2.4	< Q.L	70.50	1.09	0.19	1208
15	167	420	13.2	0.21	1.40	2.0	48.3	64.50	0.87	0.29	1552
16	224	300	11.5	0.13	1.11	1.6	24.9	120.20	3.68	0.28	882
17	208	220	10.3	0.20	0.76	2.4	62.0	76.40	1.15	0.41	1083
19	209	365	14.6	0.21	1.92	2.3	51.8	75.05	0.49	0.31	789.5
21	136	390	12.4	0.16	1.70	3.3	< Q.L	89.30	1.06	0.29	2741

Table 1 (continued)

ID	HCO_3^-	NO_3^-	SiO_2	F^-	Br^-	Oxi	Li^+	B	Cr	As	Sr^{2+}
22	195	410	15.6	0.22	0.80	1.6	50.7	117.20	9.88	0.26	1796
23	214	200	10.9	0.09	0.84	1.8	< Q.L	126.70	3.65	0.04	408
24	193	151	14.2	0.23	0.31	0.5	53.0	88.95	0.94	0.33	1413
25	228	415	16.3	0.21	1.01	1.3	74.5	108.10	1.33	0.22	1530
26	254	222	15.7	0.24	0.40	0.9	175.0	81.85	0.62	0.28	3245
27	198	505	14.7	0.24	1.34	1.4	155.0	76.95	0.81	0.45	3115
28	213	450	15.5	0.25	0.85	1.0	37.1	64.80	0.76	0.20	3066
29	220	410	15.3	0.36	1.10	1.4	10.5	110.90	0.16	0.15	1674
30	226	345	20.2	0.24	1.34	1.9	65.0	80.75	0.44	0.56	1823
31	152	420	16.6	0.21	0.89	1.3	< Q.L	48.10	0.26	0.16	2328
32	72	390	11.4	0.18	1.34	4.0	< Q.L	46.80	0.28	0.32	910
35	271	220	16.5	0.26	1.95	2.8	137.0	94.70	0.50	0.17	5095
36	246	345	15.2	0.30	1.29	1.3	138.0	71.00	0.60	0.20	3137
37	226	340	16.2	0.24	1.08	1.5	8.8	41.30	0.99	0.24	1246

Unit of EC is μS/cm

Unit of T is ºC

Unit of Eh is mV

Units of Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^- and SO_4^{2-} are mg/L

Units of HCO_3^-, NO_3^-, SiO_2, Br^-, F^- and Oxi. are mg/L

Units of Li^+, B, Sr^{2+}, As and Cr are μg/L

Q.L quantification limit

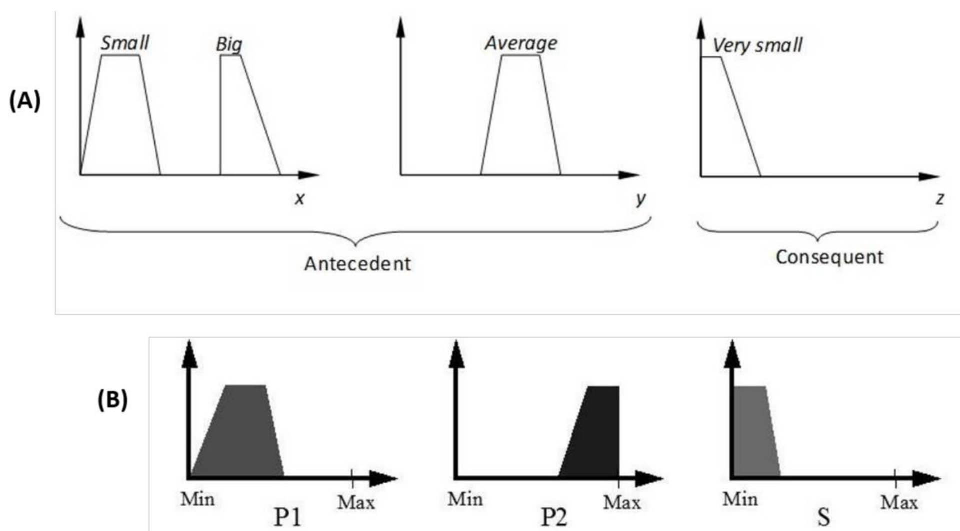

Fig. 3 **A** Example of If–Then fuzzy rule, **B** Example of fuzzy rule generated by PreFuRGe

assume the role of consequents, and therefore, the rest of the parameters will be the antecedents (Fig. 3A).

Then, if the minimum distances between data are considered and they are compared with a standard value, the variables can be grouped by using the Fuzzy C-Means algorithm (Eq. 1) in the clusters. Finally, once the previous described values are determined, the graphical fuzzy rules are generated (Fig. 3A).

$$J(X;U,V) = \sum_{i=1}^{c} \sum_{k=1}^{n} \left(\mu_{ik}\right)^m \|x_k - v_i\|_A^2 \quad (1)$$

U: fuzzy partition matrix of X. V: vector that is used to determine the cluster. m: coefficient that measures the degree of concordance of the resulting groups, μ_{ik} can take the values $0 \le \mu_{ik} \le 1$, $\|k_x - v_i\|^2_A$: is $(x_k - v_i)$ T A $(x_k - v_i)$ and is used for measuring distances.

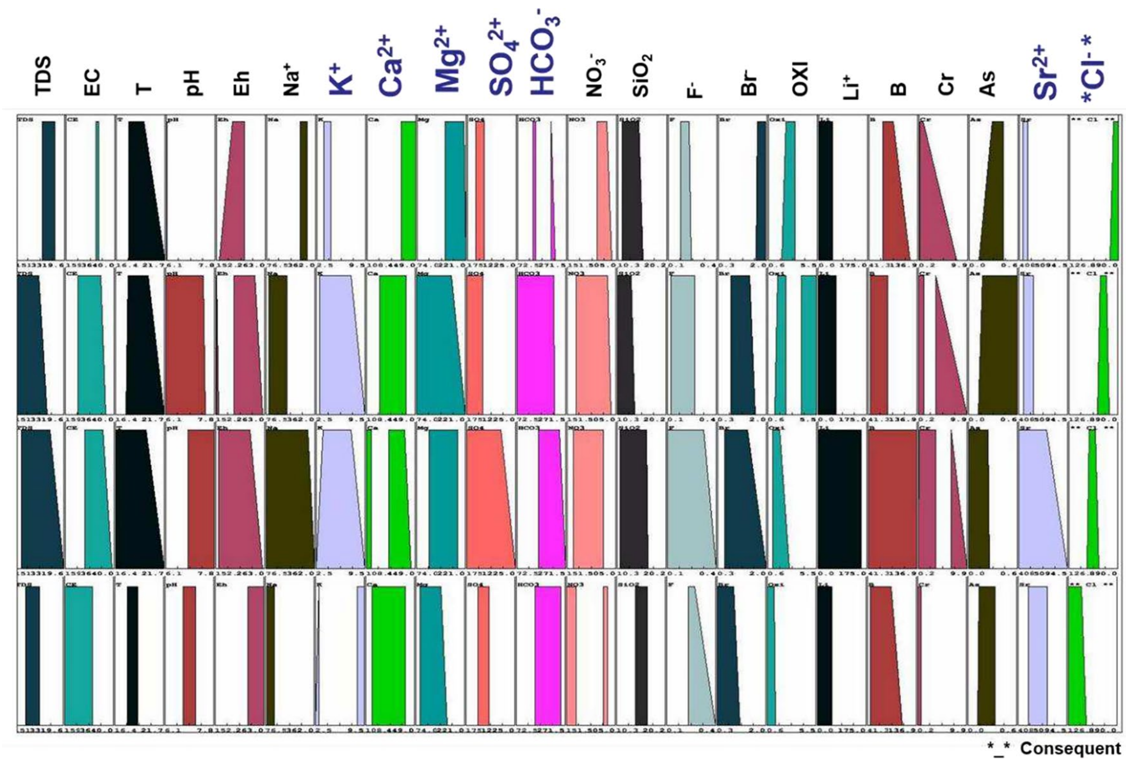

Fig. 4 Graphic fuzzy rules describing the qualitative behaviour of total Cl^-

PreFuRGe software (Aroba 2003; Davila et al. 2021) analyses and classifies the provided dataset based on the above equation, so that each selected parameter as consequent, is fuzzy clustered in an optimum number of fuzzy clusters into which the consequent under study should be partitioned (Fukuyama and Sugeno 1989). Then, each obtained fuzzy cluster is projected onto the antecedent space (Sugeno and Yasukawa 1993), and the membership grade of the antecedents to each fuzzy cluster is determined. This calculation (optimum number of fuzzy clusters) is based on the theory proposed by Fukuyama and Sugeno (1989), in which “an optimal number of fuzzy clusters are obtained by considering the variance of the data in a cluster and that of the clusters themselves”.

Finally, considering the membership functions obtained in the fuzzy clustering and fuzzy projection stages, the graphical fuzzy rules are generated (Fig. 3B). The number of rows (fuzzy rules) in each figure is determined by this optimal number of clusters, which is determined for each consequent under study.

The graphic output provided by PreFuRGe (Fig. 3B), enables an easy interpretation of the fuzzy rules in a natural language. In the generated fuzzy rules, the fuzzy set assigned to each parameter is represented by a trapezium, the parameter values are represented on the *X* axis of each fuzzy set, and the membership grade to a cluster on the *Y* axis. In the fuzzy rule presented in Fig. 3B, the parameters of the antecedent are: P1 and P2, and the consequent is S. This fuzzy rule would be interpreted as follows:

IF P1 is bigger than small and lower than average AND P2 is big THEN S is small.

In this paper, given the hydrogeochemical characteristics of the study area, the selected parameters to study as consequent in the fuzzy rules, are: chloride, strontium, sulphate, and nitrates, as they are indicators of the processes involved.

Results and discussion

Figure 4 presents the qualitative behaviour of chloride, with the Cl^- ion as the consequent of the fuzzy rules. In these rules, it should be noted that the behaviour of the K^+ is also contrary to that of Cl^-, which is compatible with the non-marine origin of K^+. According to Morell et al. (1996), the presence of K^+ in this sector of the Plana de Castellón could be related to cation exchange with Ca^{2+}, although it could also be related to the contribution of sulphate water of regional flow from the boundary aquifers (MS and Triassic units of Sierra de Espadán), which have been enriched in K^+ (Giménez-Forcada and Vega-Alegre 2015a, b).

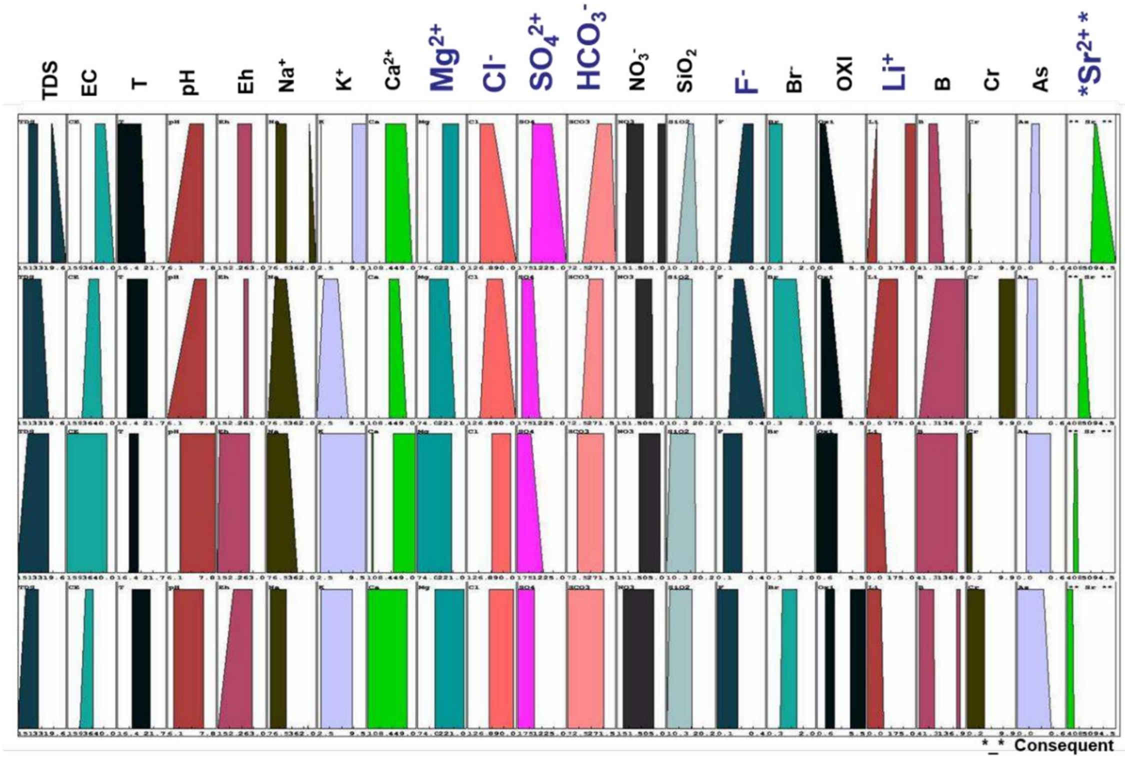

Fig. 5 Graphic fuzzy rules describing the qualitative behaviour of total Sr^{2+}

As expected, the trends of SO_4^{2-} and HCO_3^- are also the inverse of that of Cl^- (Fig. 4), although with a much less distinct component. The SO_4^{2-} and HCO_3^- ions are continental in origin, while Cl^- is of marine origin. The source of SO_4^{2-} is the dissolution of the Triassic units of the Sierra de Espadán and Mesozoic substratum (boundary aquifers).

The behaviour of Mg^{2+} is more complex (Fig. 4). Following the criteria that govern the initial conceptual model of the study area, the presence of Mg^{2+} should follow an inverse trend to Cl^-, and would be explained by the incongruent dissolution of the dolomites of the Muschelkalk (Renau-Llorens 2010). However, in this study, the behaviour of Mg^{2+} versus Cl^- is similar. The explanation for this behaviour may be related to the fact that the study area has been subject to seawater intrusion for decades (Giménez and Morell 1990, 1991, 1992; Escrig et al. 1993; Fidelibus et al. 1993; Giménez 1994; Morell et al. 2008; Renau-Llorens 2010; Ballesteros et al. 2016). Because of the intensive extraction, groundwater used for irrigation in the study area is of poor quality (electrical conductivities of up to 2750–3500 µS/cm). Farmers have been using this water, enriched in Mg^{2+}, for many years. The irrigation surplus return to groundwater (return water), being a cyclic process, the groundwater becomes enriched in the components that the groundwater initially had, and which are not absorbed or retained by plants or the soil (Renau-Llorens 2010).

The Cl^- and Sr^{2+} ions show a practically inverse relationship (Fig. 4); very low concentrations of Sr^{2+} occur when Cl^- concentrations are very high. However, very low Cl^- concentrations are only found when the concentrations of Sr^{2+} are medium to low. This relationship is not compatible with the fact that Cl^- is clearly of marine origin, hypothetically Sr^{2+} can be of marine origin but can also be of continental origin from lateral groundwater inflow (Renau-Llorens 2010; Giménez-Forcada and Vega-Alegre 2015a, b). For the continental Sr^{2+}, there are two possible origins: (1) the dissolution of the Keuper evaporites, and (2) the dissolution of the Muschelkalk dolomites (Pulido-Bosch et al. 1995), although the dissolution of dolomite may play only a minor role. The Sr^{2+} ion has been frequently used as an indicator of seawater intrusion (Morell et al. 1996). Therefore, to understand better the behaviour of Sr^{2+} and to obtain concise conclusions, we must study the Sr^{2+} as a consequent.

Figure 5 shows the qualitative behaviour of strontium, using Sr^{2+} ion as the consequent of the fuzzy rules. Comparing the Sr^{2+} tendencies with the rest of variables (ions and parameters), it is observed that Sr^{2+} has the same tendency as Mg^{2+}, Li^+, F^-, SO_4^{2-}, and HCO_3^-. In the case of Sr^{2+}, Mg^{2+}, and Li^+, this relationship has been previously demonstrated by Morell et al. (2008) using factorial analysis and by Giménez-Forcada and Vega-Alegre (2015a, b), using hierarchical agglomerative cluster analysis and principal

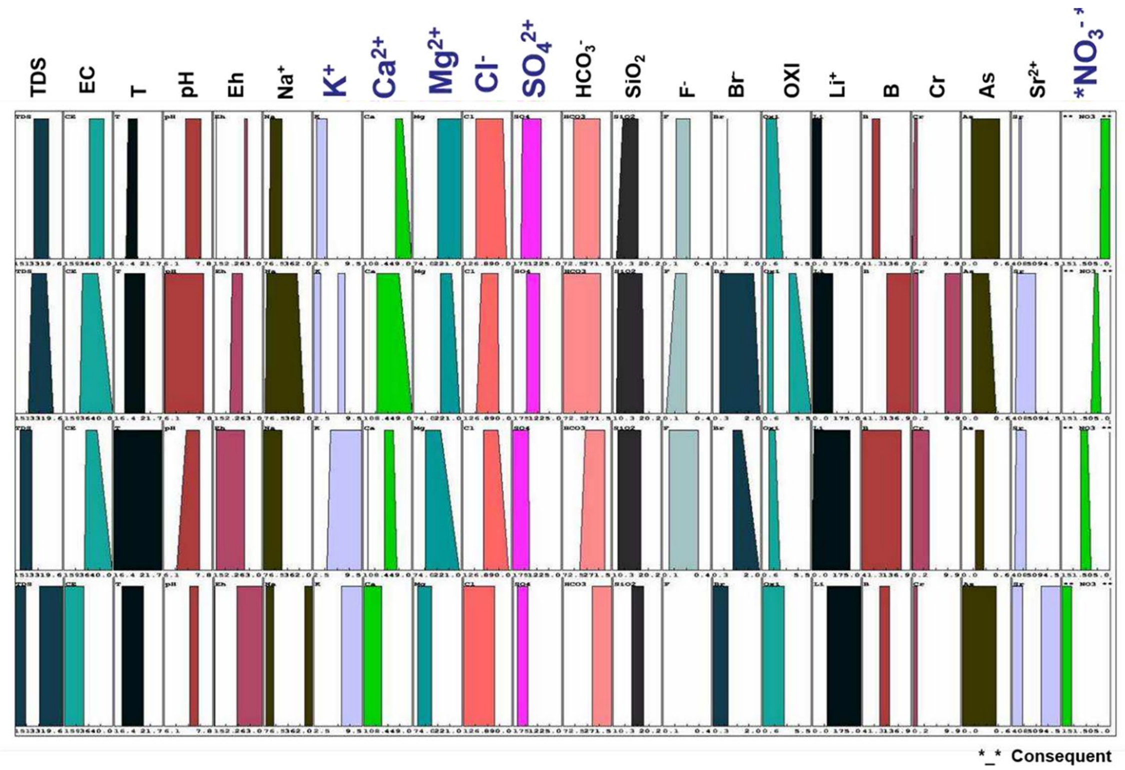

Fig. 6 Graphic fuzzy rules describing the qualitative behaviour of total NO_3^-

component analysis. This showed that these three ions did not indicate salinity caused by seawater intrusion, but that salinity was caused by continental sulphate-rich water. This also accounts for the relationship with the SO_4^{2-} component. The HCO_3^- originates from the dissolution of dolomites (Muschelkalk) and is therefore another indicator of the continental origin of groundwater. In contrast, Sr^{2+} presents a tendency opposite to that of Cl^-, which corroborates the initial conceptual model of the study area (continental origin).

In Fig. 6, it's presented the qualitative behaviour of nitrates, with NO_3^- as the consequent of the fuzzy rules. It is observed that it has the same tendency as SO_4^{2-}, Mg^{2+}, and Ca^{2+} and an opposite tendency to K^+. The NO_3^- in the study area is clearly of anthropogenic origin (contamination from intensive citrus agriculture). Decades of intensive agriculture in the area has resulted in groundwater having a very high concentration (150–450 mg/l) of NO_3^- (Renau-Llorens 2010). Although NO_3^- is of anthropic origin, its behaviour is very similar to that of Cl^- (Fig. 6). This similarity could be explained by the direct relationship between the application of fertilisers and the extraction of groundwater for irrigation. Agricultural activity, on the one hand, causes the entry of NO_3^- into the aquifer due to the use of fertilizers and, on the other hand, this activity is linked to the intensive pumping of groundwater, which causes seawater intrusion that would be characterized by the presence of Cl^- (Morell et al. 2008). Therefore, both ions (nitrates and chlorides) overlap in the same place. On the other hand, the thin thickness of the vadose zone, near the coast, makes it easier for the NO_3^- to reach the groundwater in a relatively short time since it must travel a short distance. Also, increases in NO_3^- in groundwater in some parts of the study area could also be a result of irrigation with effluent from the Vall de Uxó wastewater treatment plant (Renau-Pruñonosa et al. 2020).

NO_3^- presents behaviour similar to that of Ca^{2+} and Mg^{2+} (Fig. 6). Morell et al. (2008) have already been demonstrated this relationship by using factorial analysis and indicated that over-fertilization has been practiced in the area. Over-fertilization primarily involved nitrogen fertilizers (usually ammonium sulphate and ammonium nitrate) and has also facilitated the processes of cation exchange between NH_4^+ and Ca^{2+}. That process would also explain the direct relationship that NO_3^- can have with SO_4^{2-}. In comparison, Mg^{2+} presents the same behaviour because of its relationship with Ca^{2+}, and its origins from the uneven dissolution of dolomite (Renau-Llorens 2010).

Finally, Fig. 7 shows the qualitative behaviour of sulphate, using SO_4^{2-} as the consequent of the fuzzy rules. The behaviour of Sr^{2+}, B, Li^+, and HCO_3^- is similar to that of SO_4^{2-}, which suggests the same continental origin, and is associated with the groundwater inflows from the Triassic units. This hypothesis concurs with the findings of Renau-Llorens

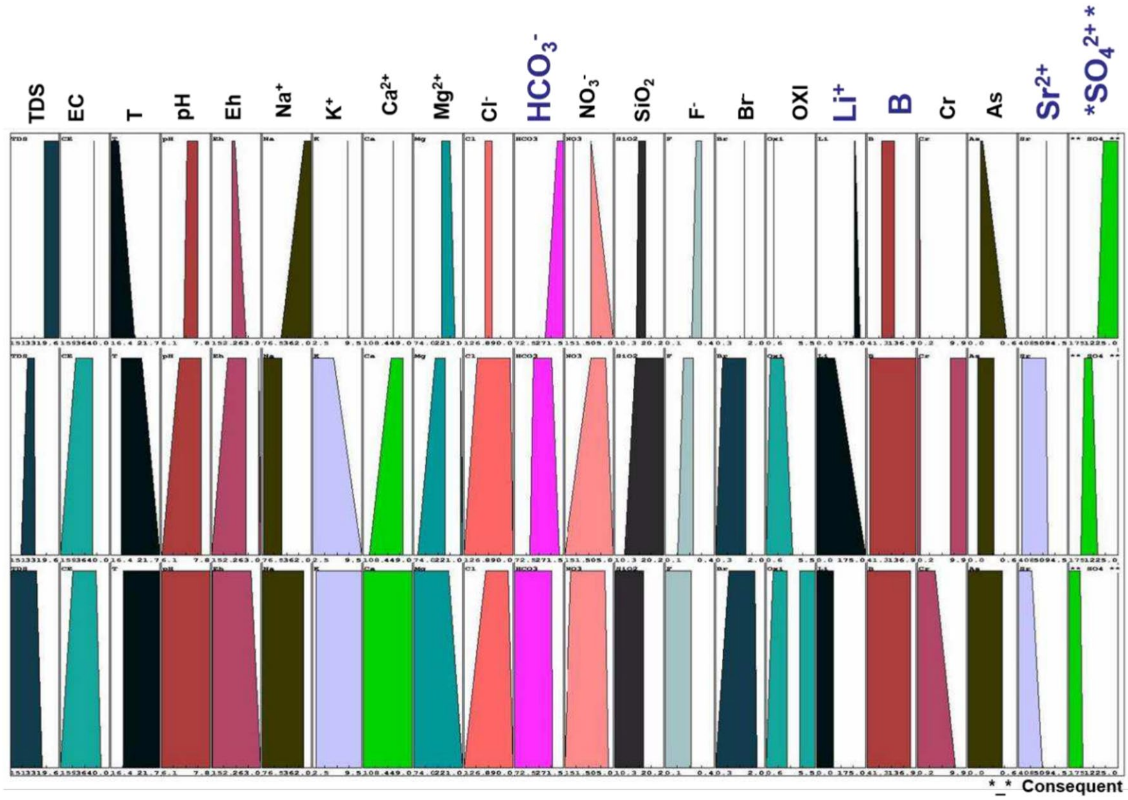

Fig. 7 Graphic fuzzy rules describing the qualitative behaviour of total SO_4^{2-}

(2010) and Giménez-Forcada and Vega-Alegre, (2015a, b), who identified the relationship between these ions using correlation analysis and ionic relationships in the southern sector of the Plana de Castellón aquifer system.

Conclusions

This paper presents a new approach to differentiate between hydrogeological processes that take place in coastal aquifers, by using a Fuzzy Logic and data mining based methodology. Although it has been rarely used in this context, it is confirmed operational models that had been previously proposed using classical statistics methods. The proposed fuzzy approach improves the efficiency for qualitative diagnosis, and also, it can be used to establish cause-effect relationships that, in contrast with classical statistical treatments, made understanding the processes involved easier.

In the obtained fuzzy models, it is demonstrated that the Sr^{2+} has two origins, marine and continental. In the first case, the $Sr^{2+}$is related to the seawater intrusion (Cl^-). On the other hand, the continental origin involves lateral groundwater inflow from boundary aquifers. In addition, the relationship of Sr^{2+} with Mg^{2+} and Li^+ has been proven.

Also, it's observed in the proposed qualitative models that the K^+ behaviour is not compatible with a marine origin and may be related to cation exchange with the Ca^{2+} ion. However, this ion could also be related, to a lesser (but not insignificant) extent, with the sulphate-rich water from the boundary aquifers, and hence with SO_4^{2-}.

In the case of Mg^{2+}, it is observed that the pattern is similar to the behaviour of Cl^-. It might be caused by intensive extraction drawing in additional sources of irrigation water loaded with Mg^{2+}. Hence, the relationship between Cl^- and Mg^{2+} is related to NO_3^-, presents a similar behaviour to Cl^-, meaning that both ions have an anthropogenic origin, being a direct relationship between fertilization and pumping in this agricultural area. Increase extraction of groundwater implies the advance of seawater intrusion and the increase of Cl^-, and the use of fertilizers induces the increase of NO_3^-.

Finally, the behaviour of Sr^{2+}, B, Li^+, and HCO_3^- is the same as that of SO_4^{2-}, which suggests the same continental origin (boundary aquifers).

This methodology based on Fuzzy Logic and data mining based could be applied in other hydrogeological environments as support for hydrogeochemistry studies. One could apply, for example, in aquifers where mixtures of recharge waters from different origins are produced.

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Author contributions The tasks performed by each team member are specified below. A.Renau-Pruñonosa coordinated the tasks to carry out the paper. A.Renau-Pruñonosa and O. Garcia-Menéndez wrote the main manuscript text. J. Aroba used the mining computer tool Predictive Fuzzy Rules Generator (PreFuRGe). O. García-Menéndez preparing figures. MV Esteller reviewed the main manuscript. MV Esteller and JA Grande helped us to understand the utility of Fuzzy Logic to hydrogeochemistry. ML de la Torre reviewed the main manuscript. I Morell and BJ Ballesteros were research group supervisor. All authors commented all versions of the manuscript, reviewed the last version and approved it.

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Declarations

Conflict of interest The authors declare no competing interests.

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