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Seawater intrusion and associated processes in a small coastal complex aquifer (Castell de Ferro, Spain)

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Abstract

An attempt to identify the hydrogeochemical processes that accompany current or past intrusion of seawater in the Castell de Ferro coastal aquifer (SE Spain) was made using ionic deltas, ionic ratios and saturation indices. Alluvial and coastal Quaternary sediments and Triassic marbles constitute this complex aquifer. Cation exchange, dolomitization, and calcite dissolution were identified as operating in the aquifer. The cation exchange processes in the detrital materials are different from those in the marbles. In addition, it was determined that while the processes of marine intrusion associated with periods of low groundwater levels still persist in the detrital aquifer, in the carbonate sector the flushing process characteristic of wetter conditions has begun.

1. Introduction

The Mediterranean coastal region of southern Spain has undergone a startling economic development over the last 20 a. This is due not only to the long-established tourist industry but also to changes in the pattern of agriculture in which traditional farming has given way to greenhouse cultivation. Because of the high profitability of this method of agriculture, the surface area under cultivation has increased notably and so has the water demand. The main source of water in the area is from groundwater and in many cases the abstractions have led to aquifer overexploitation and seawater intrusion (Benavente, 1985). The economy of the region depends to a large extent on the availability of water of adequate quality for crop irrigation, so that marine intrusion and salinization could endanger the economic future of the coastal areas.

The Castell de Ferro aquifer is located on the southeastern coast of Spain. It is bounded on all sides by mountainous relief, except to the south, where the boundary is formed by the Mediterranean Sea. It is a detrital aquifer made up of sediments from the 2 *ramblas* (ephemeral stream beds) that feed it. The aquifer occupies nearly 3 km^2; it is elongated north-south, being almost 5 km long and having a maximum width of 700 m (Fig. 1).

The mean annual temperature in this area is 18.9 °C. The mean monthly minimum temperature occurs in January (9.2 °C) and the mean monthly maximum in August (30.8 °C). Most of the precipitation occurs during November, December and January (almost 50% of annual rainfall). The driest months are July and August (less than 1%). The annual average rainfall, in the rain gauge station of Motril, is 384 mm, nevertheless the amplitude of variation is very large, between 746 and 148 mm/a (period 1985/86–1997/98). The recharge period starts in October. The mean annual potential evapotranspiration (940 mm) is much higher than the precipitation (384 mm; Pulido-Leboeuf, 2002).

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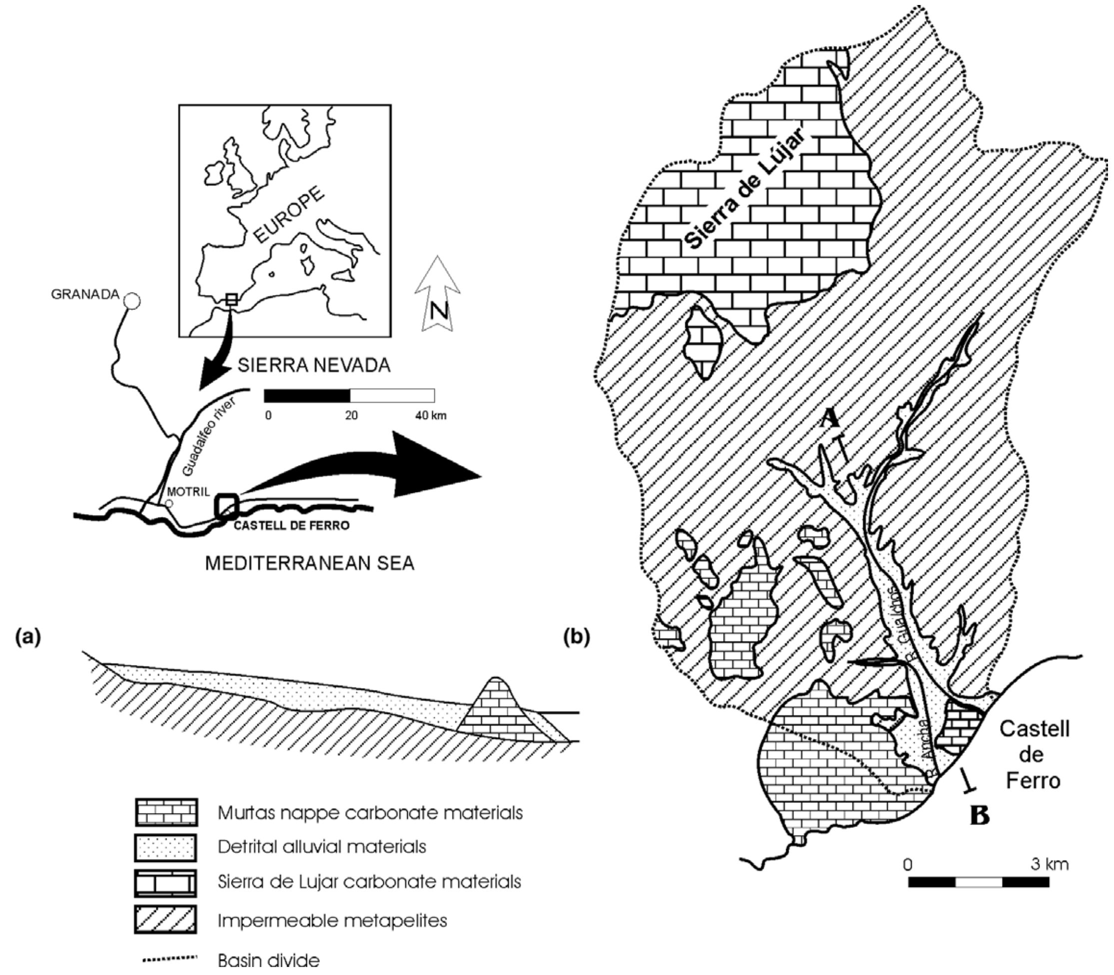

Fig. 1. Geographical and geological situation of the study area.

The study area is located in the Alpujarride Domain of the Betic Cordillera, where both pre-orogenic and post-orogenic strata are present (Aldaya, 1981). The pre-orogenic rocks belong to the Alpujarride Complex and consist of Triassic marbles and Permo-Werfenian metapelites with some limestone and gypsum intercalations. The most important post-orogenic deposits are the alluvial deposits that fill the ramblas. These are composed of gravels, sands, silts and clays and, according to drilling logs, their maximum thickness is as much as 60 m around the confluence of the 2 ramblas. In addition, there are some small encrusted beds and outcrops of cemented conglomerates that constitute, respectively, fossil riverbeds and alluvial fans.

The area of the catchment basin is 79 km^2, and its highest point lies at 1824 m a.s.l. in the Sierra de Lujar. Thus, the gradient of the streams are steep: the mean gradient of the principal streams is 9.2%. Moreover, most of the catchment basin has developed over metapelitic materials, whose low permeability means that surface-water runoff is high. However, the high permeability of the streambeds themselves favours infiltration of this runoff once it reaches these watercourses.

The Castell de Ferro aquifer has suffered frequent and persistent seawater intrusion over time and its waters are highly saline (Benavente, 1985; Calvache, 1991; Calvache and Pulido-Bosch, 1994). Notwithstanding, the data interpreted in this study do not follow this general rule, since they correspond to a wet year during which the saline wedge receded.

Representative hydrochemical facies in the aquifer show a marked difference between the headwaters, where the dominant type is Ca-HCO$_3$, and the coastal strip, where a NaCl facies dominates, and this progression clearly indicates freshwater and seawater mixing (Pulido-Leboeuf, 2002).

The karstified marble outcrop that lies in contact with both the detrital aquifer and the sea (section in

Fig. 1) acts as a preferential flow path for freshwater to the sea and for seawater into the aquifer (Calvache and Pulido-Bosch, 1994). This feature means that the Castell de Ferro system can be used experimentally to compare the processes induced by seawater intrusion in the karstic and detrital aquifers, as well as the influence of one aquifer upon the other.

A hydrochemical study of marine intrusion seems simple, given that in the majority of cases the seawater represents one extreme of the freshwater–saline water mixture. However, this mixing process is superimposed by other processes (Sánchez-Martos et al., 1999), which modify the characteristics of the mixing water, due to the initial lack of equilibrium between the aquifer matrix and the mixing water. The carbonate and clay minerals behave as geochemical buffers of salinization: they are liable to dissolution and precipitation, dolomitization–dedolomitization and cation exchange, all of which act in opposition to changes induced by seawater intrusion. Along with SO_4 reduction, these processes are the major modifying phenomena of the hydrochemistry of waters salinized by seawater intrusion (Giménez, 1994; Jones et al., 1999).

The main objective of this study was to determine if the principal determinant of groundwater salinization in the part of the aquifer that lies closest to the coast is due solely to the mixing of freshwater and seawater or if there are other hydrochemical processes involved that modify the theoretical freshwater–seawater mixture. In addition, differences between the mixing processes in the heart of the carbonate rocks and in the detritic deposits were studied.

An understanding of the geochemical processes that occur along the fresh water–seawater contact is important because these affect the permeability of the rock, in carbonated aquifers, and therefore the extent of marine intrusion suffered by an aquifer. In semiarid areas, where the summer water demand is supplied almost exclusively from groundwater resources, this behaviour assumes the utmost importance because of the economic ruin that would be the result of contamination of the aquifer by salinization.

2. Methods

A total of 115 samples were taken from 7 monitoring wells at various depths over 2 different periods (July 1998 and November 1998). Three of the 7 monitoring wells tap the full thickness of the detrital deposits, whilst the other 4 tap a marble outcrop that lies in contact with the detrital aquifer and the sea (Fig. 2). Geophysical logs of these last 4 monitoring wells were made, measuring self-potential, gamma ray, caliper, 16″ and 64″ resistivity, flowmeter, electrical conductivity and water temperature. These logs gave a detailed understanding of the geological characteristics of the monitoring wells, as well as the situation with regards to the more transmissive fractures (Fig. 2).

Sampling depths were chosen after inspection of the electrical conductivity and temperature logs. Measurements of electrical conductivity, pH and temperature were made in situ. Water samples were taken using a bailer. The samples were filtered to 0.45 μm. Two aliquots were taken: one for cation and the other for anion analysis; 0.25 ml/L of HNO_3 was added to the first aliquot to prevent precipitation. The samples were refrigerated at 4 °C until determination. Bicarbonates were determined by titration in the laboratory of the Department of Hydrogeology and Analytical Chemistry at the University of Almeria. Sodium, K, Ca, Mg, Li, B and Sr were analysed by inductively coupled plasma-mass spectrometry in the laboratory of the Department of Chemical Engineering at the University of Alicante. Chloride was determined using $AgNO_3$titration. Nitrate was determined spectrophotometrically using the brucin method; SO_4 was also determined spectrophotometrically after precipitation with barium acetate; spectrophotometric determination of Ba was according to the method of reaction with chloramine T and phenol red, whilst SiO_2 was also determined spectrophotometrically using the method of reaction with ammonium molybdate/sulphuric acid (APHA-AWWA-WEB, 1992). Analyses for Li, Sr, Si, Br and B were done only on the November samples.

Saturation indices and ionic deltas were calculated to better understand the hydrogeochemical processes that take place in the aquifer. Calculation of the ionic deltas ($\mathit{\Delta}$) consists of a comparison of the actual concentration of each constituent with its theoretical concentration for a freshwater–seawater mix calculated from the Cl concentration of the sample (Fidelibus et al., 1993):

$$\Delta C_i = C_{i,\text{sample}} - C_{i,\text{mix}},$$

where ΔC_i is the ionic delta of the ion i, $C_{i,\text{sample}}$ is the measured concentration of the ion i in the sample, and $C_{i,\text{mix}}$ is the theoretical concentration of the ion i for the conservative mix of freshwater and seawater. The theoretical mix concentration was calculated by reckoning the seawater contribution from the sample Cl concentration ($C_{\text{Cl,sample}}$), the freshwater Cl concentration ($C_{\text{Cl,f}}$) and the seawater Cl concentration ($C_{\text{Cl,sea}}$):

$$f_{\text{sea}} = \frac{(C_{\text{Cl,sample}} - C_{\text{Cl,f}})}{(C_{\text{Cl,sea}} - C_{\text{Cl,f}})}.$$

This seawater contribution was used for calculating the theoretical concentration of each ion:

$$C_{i,\text{mix}} = f_{\text{sea}} \cdot C_{i,\text{sea}} + (1 - f_{\text{sea}}) \cdot C_{i,\text{f}}.$$

These calculations can be made because Cl can be considered as a conservative tracer: the only inputs are from

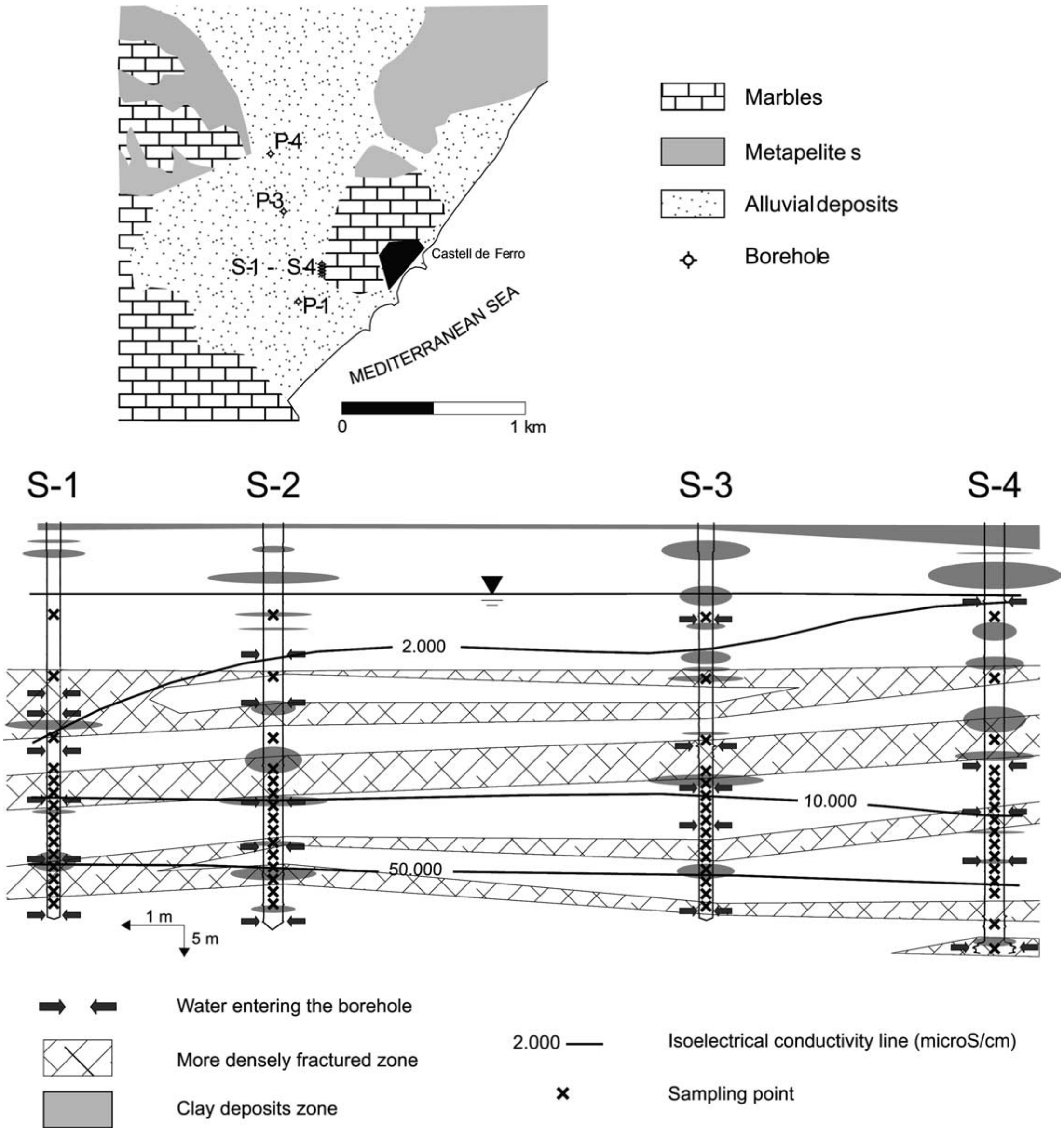

Fig. 2. Location and cross-section of sampling wells with log interpretations for the 4 monitoring wells tapping the marbles. S1 is nearest the sea.

the salts in the aquifer matrix itself or from a salinization source, and it is not usually removed from the system due to its high solubility (Appelo and Postma, 1993).

3. Results and interpretation

3.1. Water types and Piper diagram

Table 1 shows the results of the chemical analyses for each of the 7 monitoring wells sampled, as well as for a sample of seawater and another, representative of freshwater in the Castell de Ferro aquifer. This last sample was taken from a point at the beginning of the recharge area of the aquifer, where a $CaHCO_3$ water type predominates and the waters have a lower mineral content and shorter residence time.

Examination of these data shows that, for the most part, the waters of the detritic aquifer have a higher saline content, as indicated by the electrical conductivity. In the monitoring well closest to the coastline (P1) the water has a NaCl type. In general, this indicates a

Table 1
Analysis of parameters for the 7 monitoring wells sampled during November 1998

Samples	E.C.	*T*	pH	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	HCO_3^-	NO_3^-	SO_4^-	SiO_2	B	Br^-	Li^+	Sr^{2+}
P1	2660	19.8	7.54	361	28	97	90	593	408	68	200	15.1	0.2	3.9	0.03	1.3
P3	1565	19.6	7.54	333	4	73	38	223	383	60	312	24	0.2	24.5	0.02	2.5
P4	1297	20	7.34	216	13	148	52	142	387	116	309	27.2	0.2	40	0.01	1.2
S1	1100	20.2	7.54	110	18	105	45	140	345	36	120	16.7	0.1	1.3	0.02	1.1
S2	1240	20.3	7.56	162	7	71	43	172	322	40	100	15.9	0.1	0.4	0.02	1
S3	1170	21.4	7.48	155	7	71	38	157	324	36	126	14.5	0.2	1.3	0.02	1
S4	1440	21	7.49	186	18	74	40	230	325	40	135	15.4	0.1	1.9	0.01	0.9
Freshwater	643	20	–	23	2	88	42	24	340	11	105	12.1	0	0.4	0.02	0.8
Seawater	–	–	8.2	11,756	446	484	1354	21,259	157	1	2957	3	6.5	132	0.35	27.6

strong seawater influence. In this area the detrital aquifer waters also have $NaHCO_3$–SO_4 facies. In contrast, the dominant water type of the monitoring wells in the carbonate aquifer is $NaHCO_3$, becoming $CaHCO_3$ in the case of monitoring well S1. Where the seawater is most notable in the detritic aquifer is also where there is greatest evidence of Br (in particular), B, Sr and Li.

It is also obvious from these data, as commented in Section 1, that the Castell de Ferro aquifer suffered only mild marine intrusion in 1998, compared to the situation in earlier years (Benavente, 1985; Calvache, 1991), and this was due to the onset of an especially wet period over the whole of the south of Spain in 1996.

The Piper diagram in Fig. 3 shows all 115 samples as well as the theoretical freshwater–seawater line of mixing. This line was based on a sample of seawater and a sample from a well at the head of the aquifer. Most of the samples lie very close to this theoretical mixing line, with only minor deviations due to water–rock interactions. There is a small group of samples (the ones from the monitoring wells in the detrital aquifer) that deviates from the theoretical line of mixing as a result of their higher SO_4 content. The most likely source of this sulphate is from dissolution of the small amounts of gypsum scattered through the aquifer, which originate from intercalations of gypsum in the metapelitic terrains (Aldaya, 1981).

3.2. Ionic deltas

From the Piper diagram it is deduced that the most important hydrochemical process in the Castell de Ferro aquifer is the mixing of freshwater and seawater, though processes within the aquifer that augment the mineral content of the freshwater might also be important. To determine whether the salinity is derived solely from mixing with seawater, the concentrations of the various cations are presented as a function of the Cl content. Chloride is considered as a conservative ion and so represents the proportion of the mixture that is seawater.

Thus, Fig. 4 demonstrates that there is indeed enrichment of the more saline waters with respect to sodium. This is more pronounced in the July samples than in the November ones. A similar trend is shown for Ca but in this case it applies both to the freshwater as to the more saline. Potassium, on the other hand, shows the opposite tendency and is more pronounced in November than in July. Magnesium has the most distinctive pattern of distribution in the mixing water – July samples exhibit concentrations that are lower than the theoretical mixture, while November samples exceed the theoretical concentrations.

In order to determine the behaviour of these cations and identify the processes that modify the theoretical content, the calculation of the ionic delta is presented for

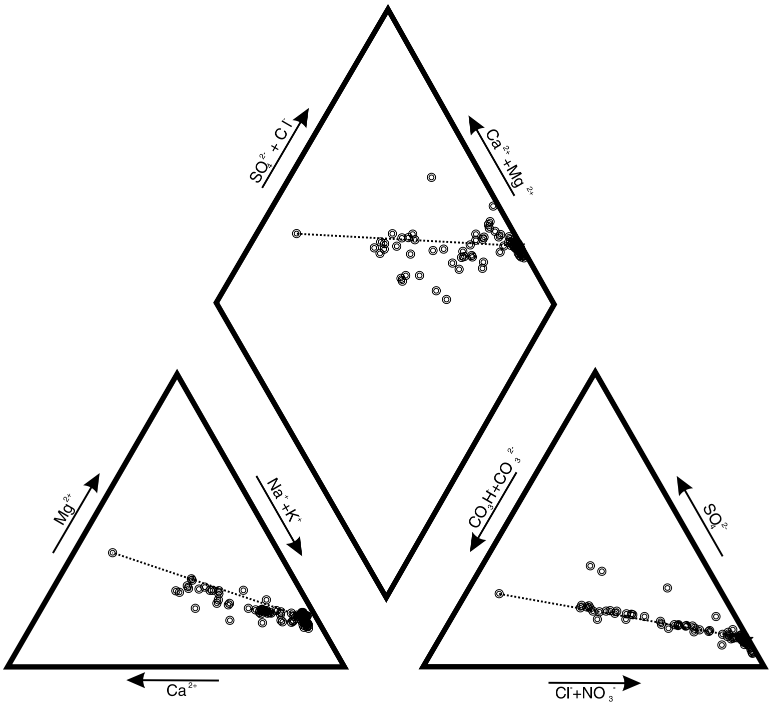

Fig. 3. Piper diagram, showing theoretical freshwater–seawater mixing line.

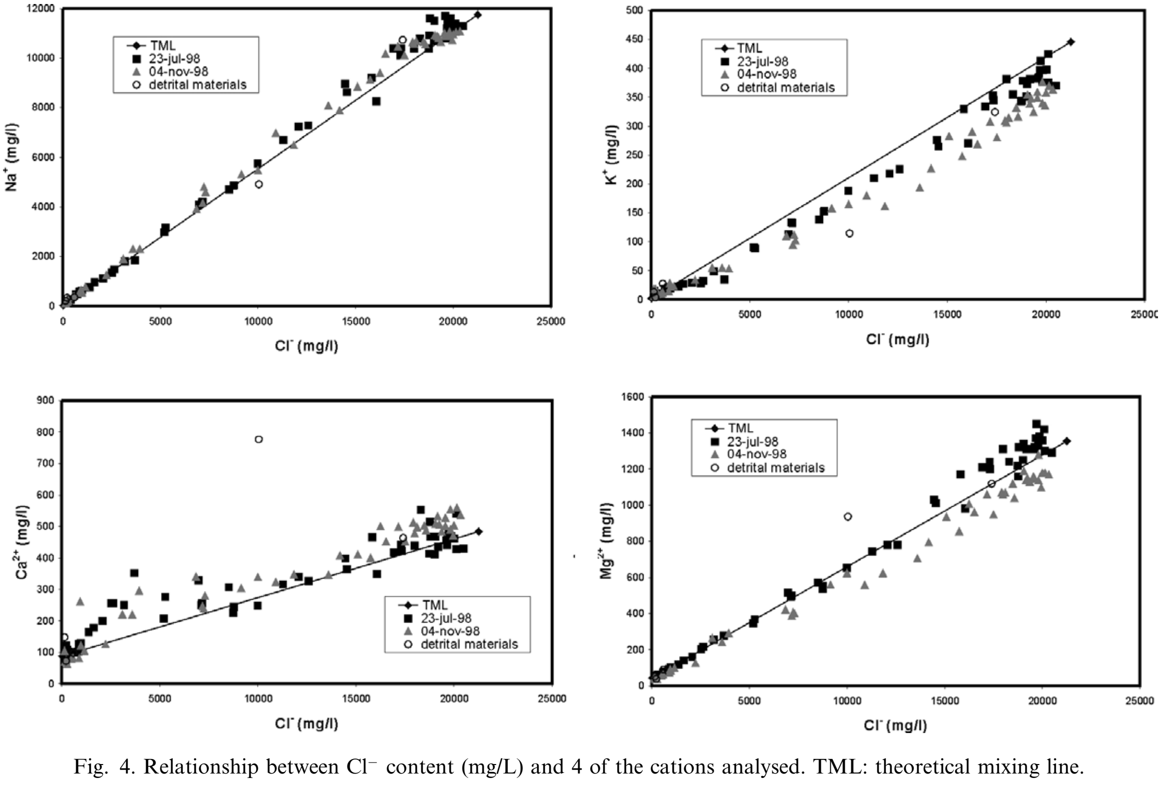

Fig. 4. Relationship between Cl^- content (mg/L) and 4 of the cations analysed. TML: theoretical mixing line.

each of the cations analysed. Fig. 5 shows the ionic deltas calculated for Na, Ca, Mg and K in the 4 monitoring wells tapping the marble (S1, 2, 3, 4), for different depths in July and November 1998.

The first thing to note is that the modifying processes in the mixing zone of this aquifer are complex and do not show a homogeneous pattern in either space or time. For example, the ΔNa^+ is usually positive but in monitoring wells S1, S3 and S4 in July and in monitoring wells S1 and S2 in November ΔNa^+ is negative – most commonly in the mixing zone and sometimes in the highly saline water (Fig. 5). The most logical explanation for this excess of Na is that a direct cation exchange is taking place between the clay present in the marble, which releases Na^+ to the solution and captures Ca^{2+} and/or Mg^{2+} from it. This process is evident for Na^+ and Mg^{2+} at points S3 and S4 and rather less so for points S1 and S2 in November. In contrast, this correspondence is not apparent in July, when there is a closer relationship between the Na and the Ca – particularly in the mixing zone – where samples with negative ΔNa^+ generally have positive ΔCa^{2+}. It is notable that cation exchange in the mixing zone of the detritic aquifer is much more significant and better defined, and produces an inverse exchange between Na and Ca–Mg.

An alternative explanation for the negative ΔMg^{2+} would be dolomitization. This may be happening at S1 and S2, where the ΔMg^{2+} is mainly negative in the mixing zone in July, but positive in the seawater fringe and always larger than the ΔCa^{2+} (which is, in some cases, negative). Such circumstances seem to be favourable to the development of dolomitization (Warren, 2000).

Potassium shows negative deltas except in the less mineralized samples, where they have a small positive value.

3.3. Saturation indices

Calcite, dolomite and gypsum saturation indices were calculated to test the possibility that SO_4-reduction and dolomitization processes are taking place. The PHREEQCI code (Charlton et al., 1997) was used to model calcite and dolomite saturation states of the

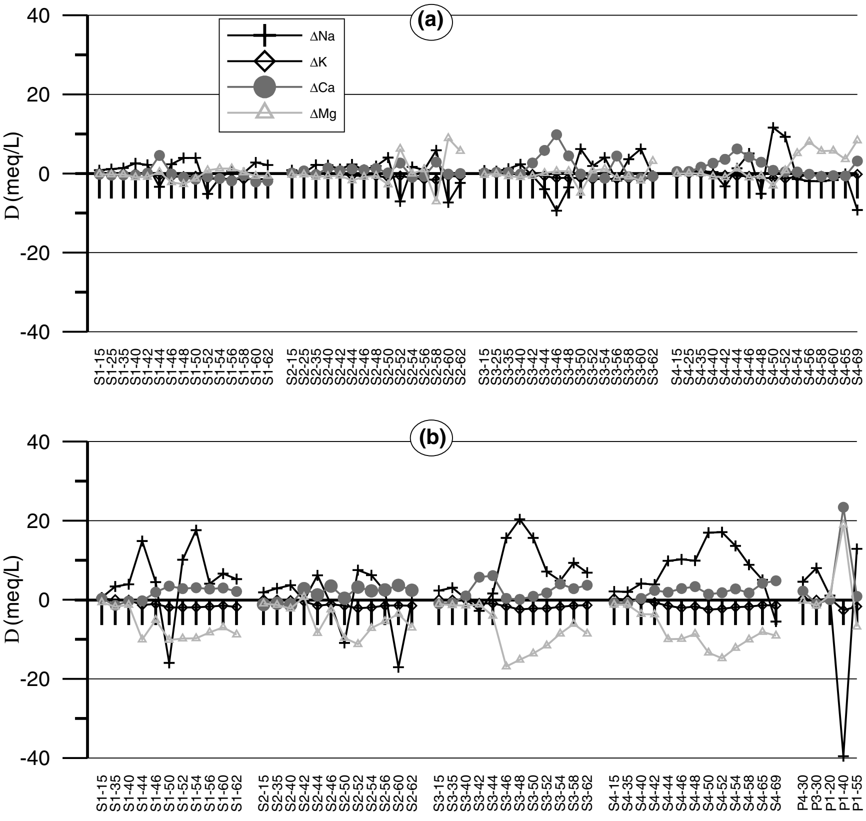

Fig. 5. Cationic deltas of the samples taken in July 1998 (a) and November 1998 (b). S1–S4 tap marbles; P wells are drilled in detrital sediments (for situation, see Fig. 2). The depth of sampling in m is indicated (e.g. S2-15; sample at 15 m in well S2).

freshwater–seawater mixing process in a closed system. Gypsum saturation indices were also modelled for different mixing rates to assess the possibility of SO_4 reduction or an alternative SO_4 source (such as gypsum dissolution).

Fig. 6 shows calcite, dolomite and gypsum saturation indices as a function of the proportion of seawater in the mixture. Also shown is the theoretical line of conservative mixing. Differences between the theoretical saturation states and the ones calculated for the samples arise due to the non-conservative nature of dissolution and precipitation of the calcite and the CO_2 flux (P_{CO_2}).

The line for saturation in calcite for conservative mixing is lowest for mixtures containing between 10% and 20% of seawater, though undersaturation does not occur (Fig. 6(a)). The July samples with less than 30% of seawater are more highly saturated than predicted, while the equivalent samples from November give saturation indices lower than predicted for conservative mixing.

The behaviour of dolomite is quite similar: samples containing more than 20% seawater are not as oversaturated as predicted for conservative mixing. The July samples with less than 20% of seawater lie above the conservative mixing line, while the November samples with less than 20% of seawater lie below this line (Fig. 6(b)). Ideal conditions for dolomitization are reached when $SI_{calcite} < 0$ and $SI_{dolomite} > 0$. Nevertheless, conditions change as a function of P_{CO_2}, and dolomitization is favoured as P_{CO_2} increases (Wigley and Plummer, 1976). Margaritz et al. (1980) stated that a basic requirement for the deposition of dolomite is for the [Mg]/[Ca] ratio to exceed 1. This condition is met in the samples containing more than 2% of seawater (Fig. 7). Curiously, samples with a seawater ratio of between 2% and 20% show an increase in this ratio from July to November, while samples with more than 20% of seawater show a decrease over the same period. The fact that several samples have positive Mg deltas and negative Ca deltas in July, and vice versa in November (Fig. 8) confirms the probability that dolomitization occurs in mixtures of more than 40% seawater, in a similar fashion to that described by Morell et al. (1997). It should not be forgotten that part of the deficit in Mg^{2+} in the November samples was explained as being due to cation exchange with Na^+, but it is also true that the more saline of the November samples yielded negative ΔNa^+ and ΔMg^{2+} in both cases. In contrast, the positive ΔCa^{2+} could point to dolomitization from July to November for these highly saline waters.

Gypsum saturation indices (Fig. 6(c)) are generally higher than the theoretical conservative mixing line, especially for mixtures containing less than 40% seawater. This suggests the existence of a SO_4 source other than seawater. Such a source could be from the dissolution of gypsum present in the metapelitic strata (although gypsum would not need to be very abundant because

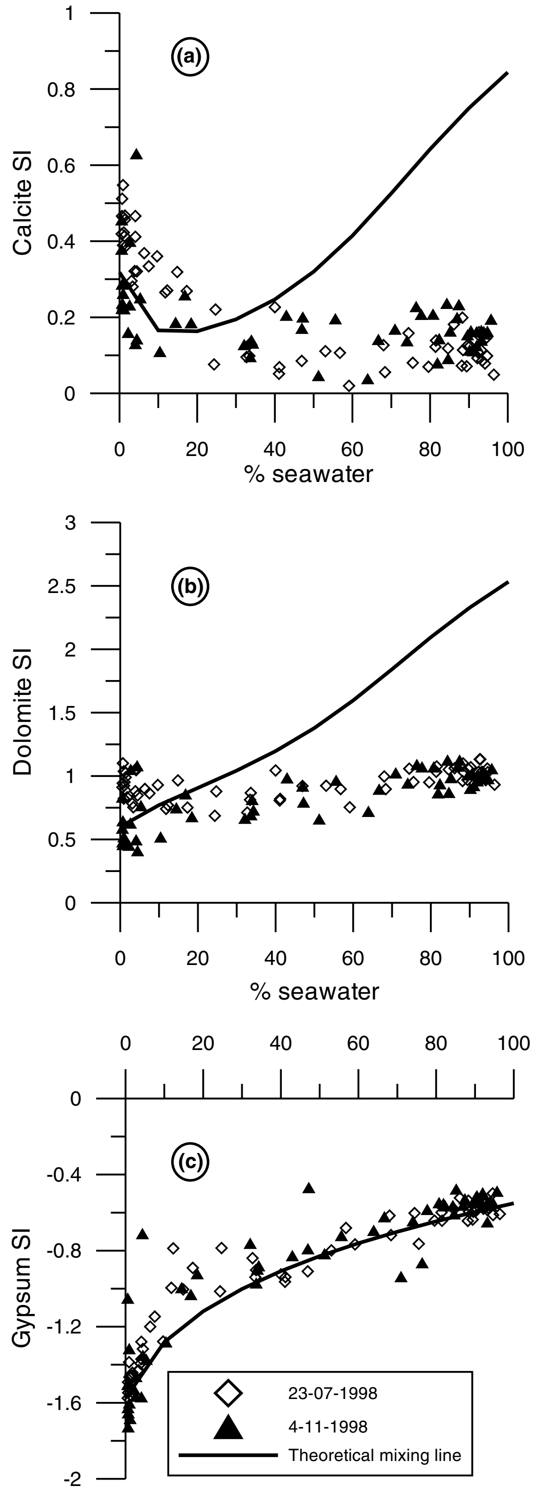

Fig. 6. Calcite (a), dolomite (b) and gypsum (c) saturation indices for the samples taken in July 1998 and November 1998. The theoretical saturation indices for conservative mixing between freshwater and seawater is also shown.

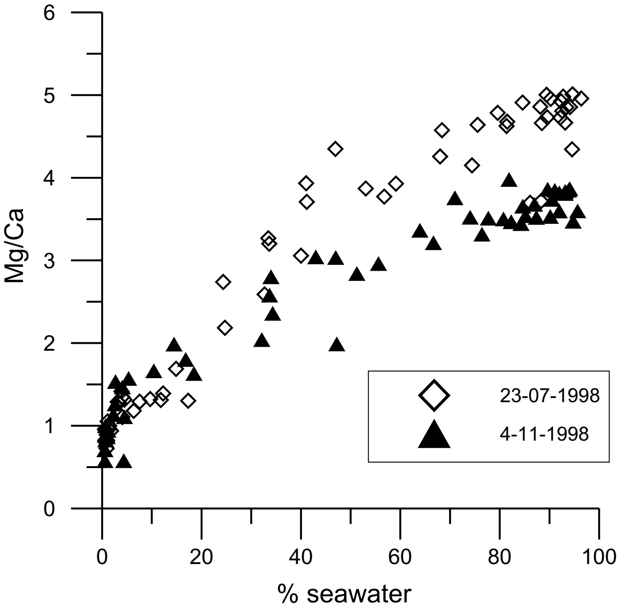

Fig. 7. Mg/Ca–% seawater plot (ion values in meq/L).

saturation is not reached). The possibility of SO_4 reduction or precipitation (Gomis-Yagües et al., 2000) in some sectors cannot be rejected, since November samples with less than 5% seawater and some of those with more than 40% seawater (both from July and November) have saturation indices well below the ones calculated for conservative mixing.

4. Conclusions

A diversity of geochemical processes is taking place in the fresh water–seawater contact zone of the Castell de Ferro aquifer, which alter the freshwater and seawater mixture away from the theoretical composition. These processes are not easily specified since they vary and are superimposed in both time and space. Perhaps the most visible process is cation exchange: this takes place between the clay and the groundwater and is quite clear in the detritic sediments as reverse exchange (removal of Na^+ from solution) of between Na^+ and Ca^{2+}–Mg^{2+} ions. Likewise, there is direct exchange (release of Na^+ to solution) between Na^+ and Mg^{2+} during the same period (November) in the carbonate outcrop. This feature is very important, as other authors have noted (Fidelibus et al., 1993; Giménez, 1994; Fidelibus, 2003), since in a coastal aquifer direct exchange is indicative of flushing, whilst reverse exchange is evidence of active marine intrusion. Therefore, even though the salinity of the water does not demonstrate it particularly well, one can say that the flushing process expected as a result of the rainy period had not yet begun in the detritic sediments at the time of sampling in 1998. Meanwhile, in the carbonate aquifer the flushing process had begun and the reason for this is that this aquifer acts as a preferential flowpath for the groundwater (Calvache and Pulido-Bosch, 1994). In the mixing zone a reverse exchange is also seen during both sampling periods, in this case between Na^+ and Ca^{2+}.

Other processes that might be taking place in the Castell de Ferro aquifer (but which are not as clear as the cation exchange) include dolomitization. Dolomitization would always appear in waters containing less than 40% seawater in November, and would be indicated by a Mg/Ca ratio (in meq/L) of more than 1, negative ΔMg^{2+} and positive ΔCa^{2+}: this combination of characteristics could not be explained by ion exchange.

In summary, the geophysical processes taking place in the fresh water-seawater mixing zone are complex. Nevertheless, it can be stated that cation exchange tends to be the dominant process in the detritic deposits, whilst the processes in the carbonate aquifer are more varied. In addition, in all cases, these processes are more active in the zone of mixing and in zones where the salinity is higher.

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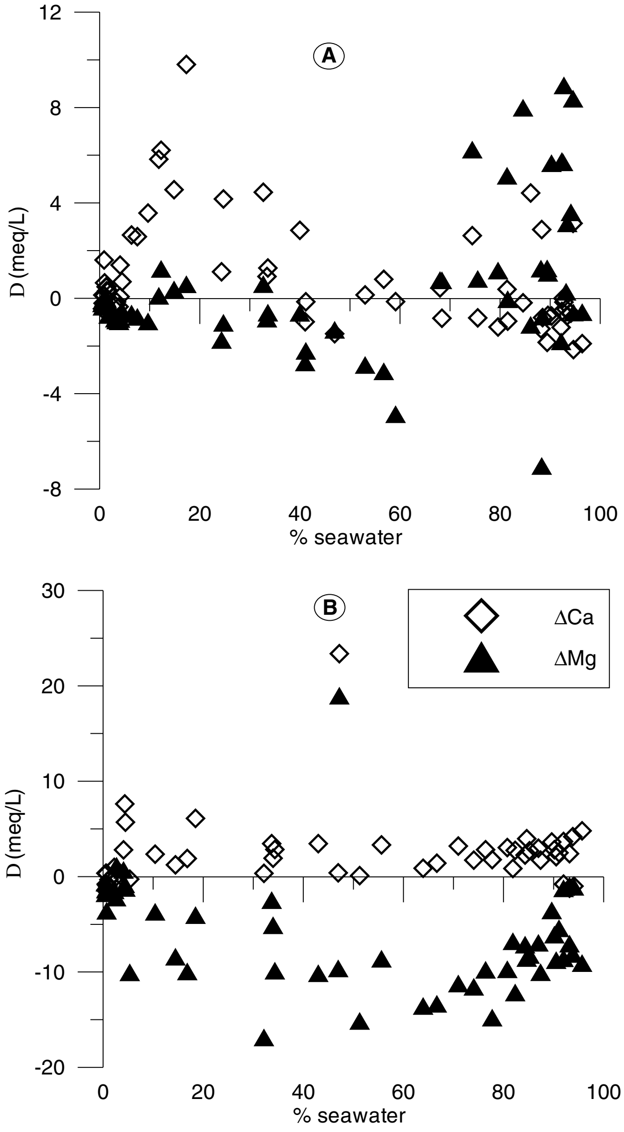

Fig. 8. ΔCa^{2+} and ΔMg^{2+}–% seawater plots for the samples taken in July 1998 (A) and November 1998 (B).

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