

Origin of boron from a complex aquifer in southeast of Spain

L. Molina · F. Sánchez-Martos · A. Pulido-Bosch · A. Vallejos

Abstract A study of the boron content alongside Cl^- , SO_4^{2-} , and NO_3^- ions has enabled the factors that determine its origin to be distinguished. In carbonate rocks, with their higher saline content, boron is associated with marine intrusion. In the Plioquaternary aquifer, its origin is complex and is associated with three processes: marine intrusion, where there is a high Cl^- content and the B/Cl^- ratio is close to that of seawater; anthropogenic pollution, at points with high B and NO_3^- concentrations and where the B/Cl^- ratio is higher than that of seawater; and the dissolution of evaporite deposits present in the Plioquaternary strata, at a series of sampling points where the $\text{SO}_4^{2-}/\text{Cl}^-$ ratio is elevated and B content is high.

Keywords Boron · Salinization · Anthropogenic pollution · Marine intrusion · Evaporites

Introduction

The Aguadulce aquifer unit is situated in the south-east of Spain, between the Sierra de Gádor and the Mediterranean Sea (Fig. 1). The oldest outcrops in the area correspond to the Alpujarride Complex, which is characterized by the superposition of the nappes of Gádor and Felix. The Gádor nappe is composed of phyllites and quartzites Permowerfenian, overlain by a thick limestone-dolomite series from the Triassic epoch, which can exceed 1000 m. The

Felix nappe has the same sequence but its thickness is around 100 m.

The post-orogenic terrains occupy the large plain at the foot of the Sierra de Gádor and correspond to deposits that represent the Upper Miocene to the Quaternary. The Miocene rocks are dolomicrites or biomicrites, marls, gypsums, volcanic rocks and conglomerates whose thickness can exceed 100 m. To the south of Vicar a volcanic agglomerate outcrops, comprised almost exclusively of orthopyroxenite andesites. The Pliocene deposits are formed by a thick, marly formation (500–1000 m), which gradually gives way to sandy marls, sands and calcarenites. The Quaternary deposits correspond to outwash cones, which are well developed at the foot of the Sierra de Gádor, and detritic deposits related to former salt flats and raised beaches (Fourniguet 1977).

The Aguadulce aquifer unit (Fig. 1) has a complex geometry as a consequence of the lithological diversity and structure of the area. The carbonate strata of the Felix nappe are highly developed and overlie the Gádor nappe, which is thicker. A layer of phyllites separates the two units, making two distinct aquifer layers. Miocene calcarenites are situated over the carbonate layers of both nappes and are in hydraulic connection with these; Pliocene calcarenites and other more recent detritic sediments can also be found, which also behave as aquifers (Fig. 1). Therefore, there would be an upper aquifer (Pliocene calcarenites together with Plioquaternary detritic materials) and two carbonate aquifers: one corresponding to the Felix carbonate strata and the other comprising the Gádor carbonate rocks, which extend to depth. Overall, there is a sequence of formations separated by layers of low permeability. The intense fracturing favours a hydraulic connection between the various aquifer levels, which, under a natural regime, would show the same piezometric level. However, overexploitation of the unit has given rise to a difference in levels, depending on the cross-section. The water in some parts is highly saline, and this has created serious problems for urban and agricultural water supplies. The aim of the current study is to discuss the source of salinity, analysing in particular the boron content of the groundwaters. The factors that control the origin of the boron and its relationship with salinity are considered, with respect to the different lithology of the aquifer materials and the variability of the processes that can develop in the Aguadulce unit.

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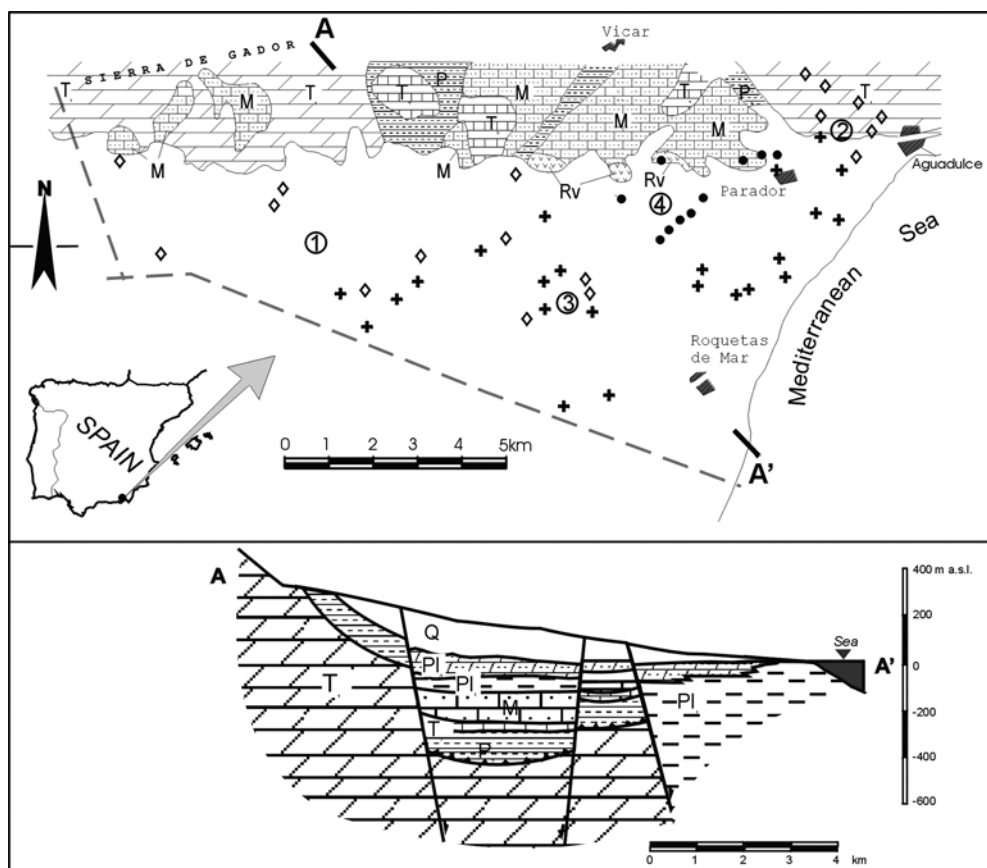


Fig. 1

Situation and geological scheme of the Aguadulce Unit. P₁: Gádor metapelites; T₁: Gádor carbonates; P₂: Felix metapelites; T₂: Felix carbonates; Rv: Conglomerates and volcanic rocks; M: Miocene calcarenites; Pl₁: Pliocene marls; Pl₂: Pliocene calcarenites; Q: Quaternary sediments. A–A': Cross-section. ① to ④: sampling sites. +: Plioquaternary aquifer, ●: Felix carbonate aquifer, ◇: Gádor carbonate aquifer

Methods

Hydrogeochemical data was obtained from a monitoring network of 55 boreholes, sampled in October 1991. The base data corresponding to measurements of temperature and electrical conductivity were determined in the field under pumping conditions. The data utilised in the present study correspond to concentrations of the anions Cl⁻, SO₄²⁻, and NO₃⁻ (Table 1). The content in these ions was determined using ion chromatography. Boron was determined using inductively coupled plasma analysis (ICP-OES).

Results

The water of the Aguadulce unit shows a gradation of facies from calcium–magnesium bicarbonate to sodium chloride. Salinity increases from the edge of the Sierra de Gádor towards the sea, with hydraulic connection in the extreme east. The water in the limestone–dolomite aquifer on the western edge of the Aguadulce unit (site 1) is the least saline and its content of Cl⁻ (<150 mg L⁻¹) and SO₄²⁻ (<70 mg L⁻¹) is the lowest of the area (Table 1). In the eastern part of the unit (site 2), the waters are more saline; Cl⁻ oscillates between 261 and 1032 mg L⁻¹ and SO₄²⁻ between 54 and 150 mg L⁻¹.

These carbonate rocks extend at depth throughout the central area of the Aguadulce unit (site 3), where they indicate a greater chemical variability. Cl⁻ varies from 20 to 768 mg L⁻¹, SO₄²⁻ from 24 to 104 mg L⁻¹, and the water temperature is the highest recorded in the area (27 °C–29 °C). The boreholes that tap the Miocene calcarenites and the dolomites of the Felix unit (site 4) are high in Cl⁻, with maximum values of 4254 mg L⁻¹. There has been considerable abstraction in this area, and values of electrical conductivity of close to 15000 μS cm⁻¹ have been recorded. Ionic concentrations in the water in the Plioquaternary levels show wide variation: Cl⁻ oscillates between 193 and 3143 mg L⁻¹, SO₄²⁻ between 59 and 563 mg L⁻¹, while the temperature doesn't overcome 23.5 °C.

The boron and nitrate content of the water reflects this hydrochemical heterogeneity. With regard to the boron, the lowest values are recorded in the Gádor limestone–dolomite (0.04 to 0.41 mg L⁻¹). Boreholes tapping the Miocene calcarenites and the dolomites of the Felix unit show a wide range (0.20–6.32 mg L⁻¹). The highest concentrations encountered in the area (13.8 and 11.7 mg L⁻¹) are recorded from boreholes that tap the Plioquaternary aquifer. These waters are highly variable: boron varies between 0.15 mg L⁻¹ and 13.8 mg L⁻¹ with a mean concentration of 1.93 mg L⁻¹. The waters from Gádor aquifer have low or null concentrations in nitrate (<11 mg L⁻¹). However the samples from the carbonate unit of Felix

Table 1

Comparison of sampling sites (see Fig. 1, *: samples distributed in the whole area), aquifer (GC: Gádor carbonate aquifer, FC: Felix carbonate aquifer, Pl: Plioquaternary aquifer) and chemical data (TDS: total dissolved solids, Thickness UZ: thickness of unsaturated zone)

No.	Site	Aquifer	T (°C)	Conduc-tivity ($\mu\text{S cm}^{-1}$)	Cl (mg L^{-1})	SO ₄ (mg L^{-1})	NO ₃ (mg L^{-1})	B (mg L^{-1})	TDS (mg L^{-1})	Thickness UZ(m)
567	1	GC	22.5	530	35	22	4.1	0.05	289	260.0
573	1	GC	22.9	780	110	39	5.8	0.17	442	250.0
683	1	GC	24.5	470	7	20	3.0	0.06	252	237.0
696	1	GC	27.7	590	39	43	3.5	0.11	335	229.0
737	1	GC	20.9	500	16	41	7.0	0.04	315	342.0
745	1	GC	22.1	730	68	45	10.6	0.17	393	171.0
748	1	GC	20.6	1000	147	67	2.4	0.19	546	188.0
802	1	GC	19.5	710	57	54	0.0	0.31	369	250.0
818	1	GC	21.2	910	142	57	11.0	0.15	543	145.0
1031	2	GC	23.7	1210	261	54	4.1	0.04	680	201.0
1032	2	GC	23.2	1700	400	120	2.3	0.09	1001	58.0
1035	2	GC	22.0	3500	984	140	10.0	0.08	1930	89.0
1037	2	GC	21.6	2200	643	140	10.5	0.41	1391	97.8
1049	2	GC	21.2	3200	919	128	10.0	0.13	1781	52.0
1054	2	GC	20.2	3200	854	100	5.0	0.14	1673	51.0
1137	2	GC	21.3	3600	1032	150	5.0	0.20	2005	90.0
839	3	GC	27.3	440	20	24	0.1	0.09	244	100.0
894	3	GC	28.8	510	38	31	0.1	0.11	277	91.0
903	3	GC	28.8	2300	768	104	9.3	0.57	1483	71.0
891	4	FC	24.0	2150	290	220	0.1	4.67	1000	98.0
963	4	FC	23.7	11000	3419	400	20.0	6.34	5998	69.5
956	4	FC	22.8	7150	2043	235	17.0	0.72	3944	70.8
889	4	FC	21.8	1950	535	150	20.0	0.26	1305	122.0
948	4	FC	20.1	1850	348	320	2.3	0.20	1177	70.5
949	4	FC	21.6	9150	3192	418	33.9	0.61	5753	72.0
950	4	FC	21.3	9600	3440	471	29.3	0.66	6196	71.0
965	4	FC	23.2	14600	4254	541	1.6	0.78	7899	75.0
900	4	FC	25.3	8300	2406	300	10.0	0.55	4435	69.8
961	4	FC	24.1	6000	1871	300	50.0	0.67	3645	69.5
962	4	FC	24.1	5300	1564	300	60.0	0.63	3174	67.8
702	*	Pl	22.5	3000	808	125	42.5	0.39	1687	225.0
752	*	Pl	23.5	1050	193	60	5.0	0.26	594	74.0
754	*	Pl	23.0	3600	909	223	69.0	0.48	1980	104.0
755	*	Pl	23.0	3150	628	361	25.6	0.18	1685	100.0
810	*	Pl	21.3	1800	445	80	24.0	0.24	1026	150.0
821	*	Pl	20.9	1350	260	59	0.1	0.17	687	95.0
821	*	Pl	20.9	1350	260	59	0.1	0.17	687	95.0
825	*	Pl	22.9	3700	1141	127	41.0	0.37	2159	109.0
828	*	Pl	22.7	2900	767	111	26.0	0.33	1591	69.0
844	*	Pl	22.8	6200	1894	310	58.3	1.01	3747	69.8
845	*	Pl	23.1	9600	3143	400	120.0	1.01	5660	62.8
859	*	Pl	19.1	4000	907	208	198.5	0.60	2187	15.0
905	*	Pl	22.5	5900	1644	377	183.0	1.14	3602	40.0
914	*	Pl	21.5	3400	896	151	0.0	2.68	1807	19.0
952	*	Pl	21.1	5000	1355	556	102.0	11.71	3145	69.5
977	*	Pl	23.3	4700	1196	376	245.0	1.54	2947	31.0
978	*	Pl	24.0	4500	1201	293	136.5	1.04	2668	30.0
980	*	Pl	23.0	6800	1934	563	213.4	1.98	4467	29.0
989	*	Pl	20.2	3650	815	364	223.0	0.76	2245	12.0
1038	*	Pl	20.6	4100	1199	172	29.0	0.15	2293	49.0
1076	*	Pl	19.8	2800	725	161	60.3	0.31	1628	45.0
1100	*	Pl	20.5	3800	897	351	114.5	4.24	2279	24.0
1106	*	Pl	19.6	3500	969	138	13.0	0.26	1948	12.0
1124	*	Pl	21.0	4800	1177	443	187.5	13.85	3129	10.0
1132	*	Pl	21.2	5500	1608	347	35.5	0.72	3256	4.0

aquifer end up reaching 60 mg L^{-1} , since many of these boreholes are sunk into intermediate and superficial formations. The waters from the modern materials show a great variability with regard to the content in this ion,

from 0 to 245 mg L^{-1} . The NO_3^- ion has been used as a tracer to demonstrate the possible existence of connections between superficial and deep aquifers in this area (Pulido–Bosch et al. 2000).

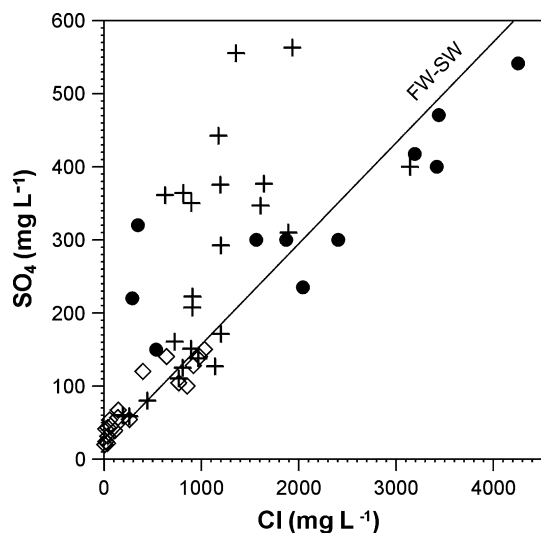


Fig. 2

SO₄/Cl ratio. +: Plioquaternary aquifer, ●: Felix carbonate aquifer, ◇: Gádor carbonate aquifer. FW–SW: mixing line freshwater–seawater

Discussion

The chemical variability is indicative of the complex origin of the salinity of the groundwater, which is due to more than one process. The graph of Cl⁻ against SO₄²⁻ content follows a model of saline enrichment (Fig. 2). The points corresponding to the boreholes that exploit the carbonate levels lie close to the theoretical line of mixing between fresh water and seawater. The points that penetrate the Gádor nappe (Cl⁻ < 1000 mg L⁻¹) are less saline than those in the Felix nappe (200 < Cl⁻ < 4300 mg L⁻¹). All these points are situated in an area where exploitation has been intense, and this has caused the piezometric levels to become negative (Molina 1998). The remaining boreholes have a much higher SO₄²⁻ content and lie above the fresh water–seawater (FW–SW) line of mixing; this indicates that inputs of SO₄²⁻ other than seawater must exist. These points exploit the Plioquaternary aquifer levels and the Felix carbonates.

Boron tends to be concentrated in environments where evaporites are present, or in brines of marine and/or continental origin (Leeman and Sisson 1996; Uhlman 1991). For this reason, there is a correlation between the salinity and the boron content in the sediments; boron has even been used in conjunction with various ionic ratios to distinguish the origin of water salinity (Arad et al. 1986; Vengosh et al. 1991; Sánchez-Martos et al. 1999). Accordingly, one could deduce that there is a direct relationship between water conductivity and its boron content. In the Aguadulce unit this relationship is unclear, given that the sampling points yielding maximum boron content (14 mg L⁻¹) have an electrical conductivity of approximately 5000 μS cm⁻¹, whilst other points exhibit the opposite situation of high salinity (9000 μS cm⁻¹) and low boron content (0.6 mg L⁻¹).

To address this question in more detail, the boron content was compared with that of Cl⁻ and SO₄²⁻, the two ions that

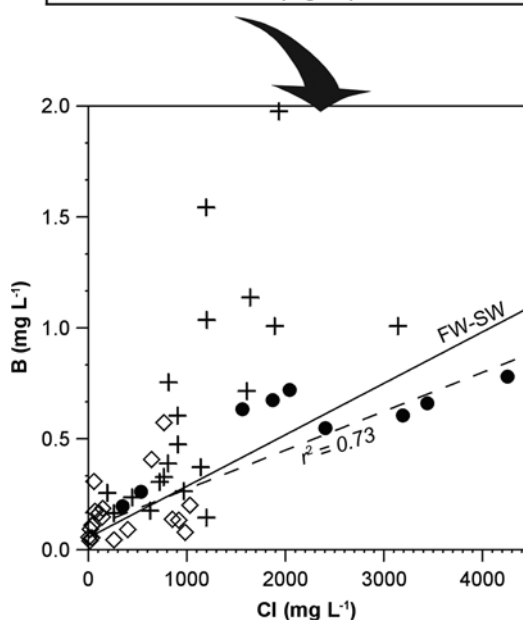
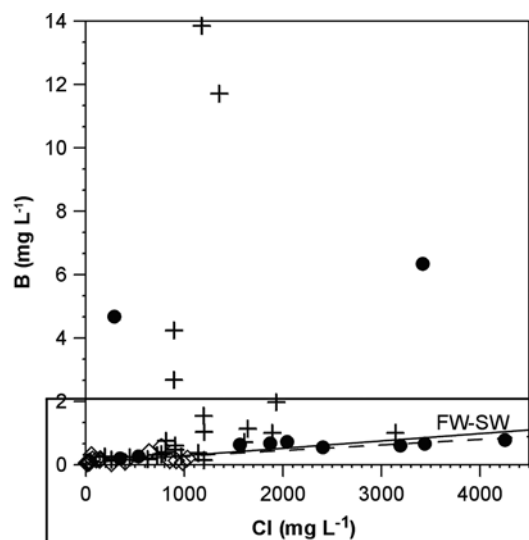


Fig. 3

B/Cl ratio. +: Plioquaternary aquifer, ●: Felix carbonate aquifer, ◇: Gádor carbonate aquifer. FW–SW: mixing line freshwater–seawater

determine the salinity of the waters. The B/Cl ratio (Fig. 3) is lowest at the sampling points tapping the carbonate rocks of the Felix and Gádor nappes, where the salt content is highest (electrical conductivity > 2000 μS cm⁻¹, and Cl⁻ > 300 mg L⁻¹). These points lie directly over the fresh water–seawater line of mixing. These materials have a carbonate lithology, and the piezometric level is negative as a consequence of the intense exploitation they have undergone (Molina 1998). Marine intrusion takes place within these strata, as demonstrated by hydrogeochemical criteria (Pulido-Bosch et al. 1991). In fact, the relatively high concentration of boron of seawater allows it to be considered as an indicator of seawater intrusion. Accordingly, there is a great similarity between the line that represents fresh water–seawater mixing and the regression line fitted to all the points that tap carbonate rocks ($r^2 = 0.73$).

Considering these two factors, the sources of boron and Cl^- must be associated with seawater. Nonetheless, other points tapping the Felix carbonate rocks do not fall close to the line of FW–SW mixing (Fig. 3). Their concentration of boron is high (4 and 6 mg L^{-1}), Cl^- content is highly variable (290 and 3400 mg L^{-1}) and their temperature elevated (23.7 and 24 $^\circ\text{C}$). Though these boreholes exploit the carbonate rocks, they cross various Miocene volcanic materials, and this may explain their high boron content and high B/Cl ratio. The high contents detected in the thermal waters are frequently associated with geothermal processes (Kakianha et al. 1989) and it may be also attributable to changes in the volcanic rocks themselves and to hydrothermal activity (Risacher and Fritz 1991; Seyfried et al. 1983), given that it is known that temperature controls liberation of B by means of water-rock interaction (Arnorsson and Andresdottir 1995).

With respect to the Plioquaternary aquifer levels, it can be seen that some points lie very close to the theoretical line of FW–SW mixing (Fig. 3). Here, the content of Cl^- is elevated ($>3000 \text{ mg L}^{-1}$) and the B/Cl ratio oscillates within a range close to that of seawater. At these points the boron may be associated with seawater intrusion, as is the case for the carbonate rocks described above. However, the majority of the points, which represent boreholes penetrating the Plioquaternary levels, are situated away from the FW–SW mixing line. Their boron concentration is high (1–5 mg L^{-1}), Cl^- concentrations vary between 500 and 3000 mg L^{-1} , and the B/Cl ratio is higher than sea-

water. This variability may be due to a combination of processes.

Thus, one must consider the limited thickness of the unsaturated zone and the intensive agricultural activity that goes on in the area (Pulido–Bosch et al. 2000), which leads to anthropogenic pollution. The boreholes tap the Plioquaternary levels in an area where the unsaturated zone is thinner, and the water contains the highest NO_3^- and B concentrations ($<100 \text{ mg L}^{-1}$ and 0.5–13 mg L^{-1} , respectively; Fig. 4). As a result, the origin of the boron is associated with the use of fertilizers as well as with point sources of pollution of underground wastewater, as highlighted by various authors (Uhlman 1991; Mirecki and Parks 1994; Leenhouts et al. 1998; Vengosh et al. 1994). Nevertheless, analysis of these processes requires a further important factor to be considered, namely the lithology of the Plioquaternary deposits. These have a marine origin: they are Pliocene calcarenites that have been deposited within enclosed basins, and so they may contain evaporite intercalations or even brines (Pulido–Bosch et al. 1989). The Plioquaternary aquifer sampling points produce elevated SO_4/Cl ratios, which lie above the theoretical FW–SW line of mixing (Fig. 2). We consider that these two ions are not directly related to processes of intrusion and that there has to be another process to explain the higher SO_4^{2-} values, probably related to the dissolution of the evaporite intercalations. The influence of these evaporites may explain the high B content in this zone, since this element has been associated with the dissolution of evaporite rocks

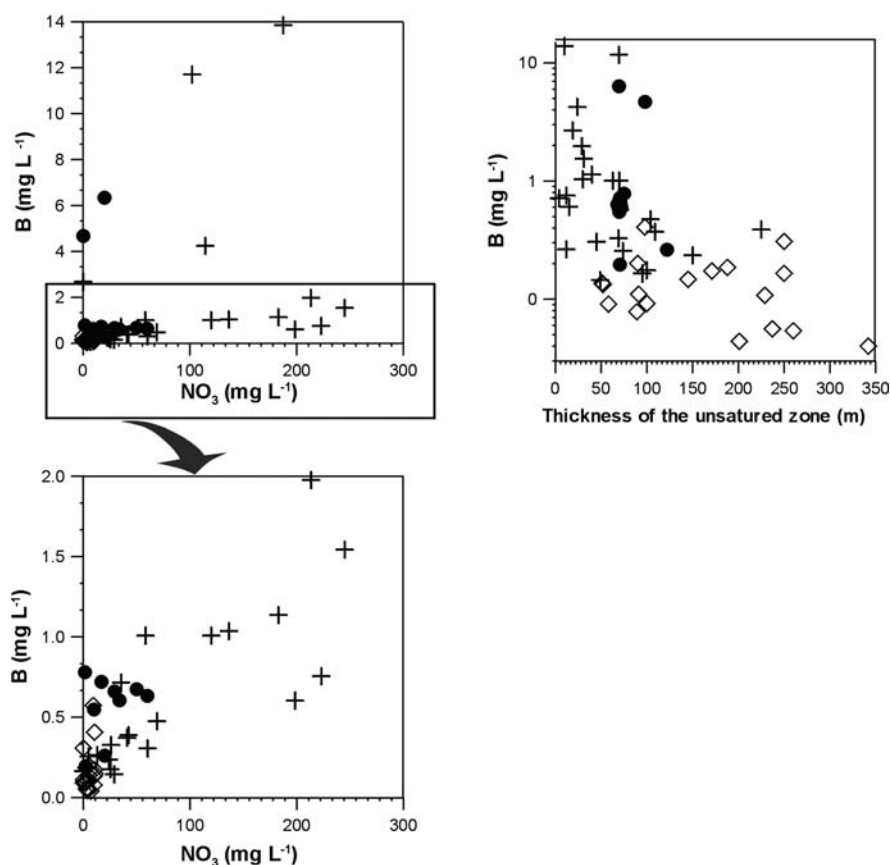


Fig. 4

B/ NO_3 ratio and B/thickness unsaturated zone. +: Plioquaternary aquifer, ●: Felix carbonate aquifer, ◇: Gádor carbonate aquifer

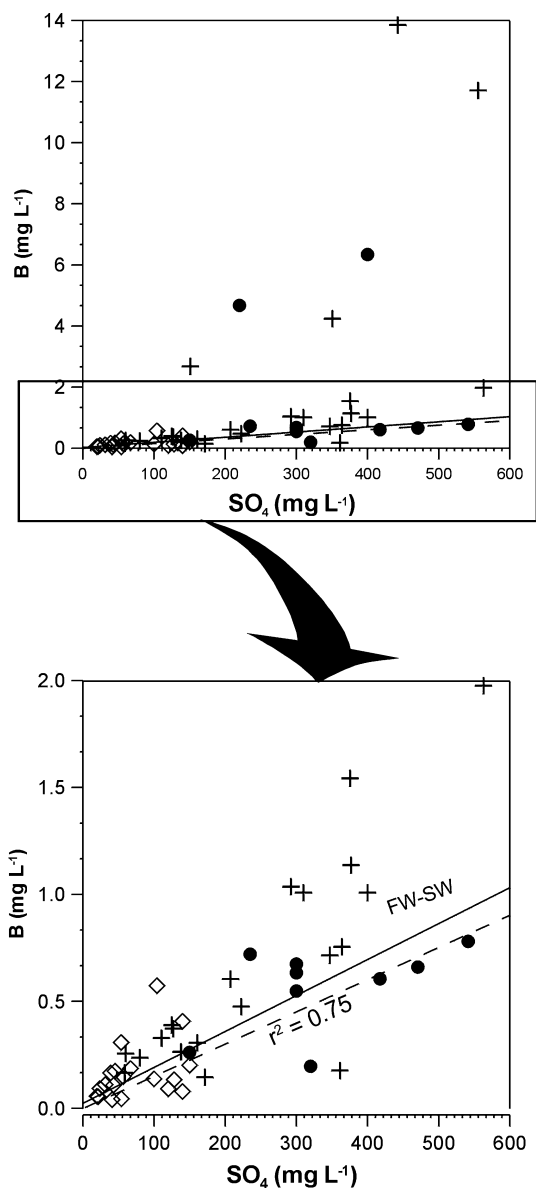


Fig. 5

B/SO₄ ratio. +: Plioquaternary aquifer, ●: Felix carbonate aquifer, ◇: Gádor carbonate aquifer. FW–SW: mixing line freshwater–seawater

(Leeman and Sisson 1996; Neal et al. 1998; Rose et al. 2000).

In order to discriminate between the anthropogenic and evaporite influences on the water composition, we considered the relationship between B and SO₄ (Fig. 5). Points with a high B content lie away from the general trend, indicating that their origin is probably different. This situation is clearly demonstrated in the Plioquaternary aquifer, where there are four points with a high B content (2–14 mg L⁻¹) and a wide variation in SO₄²⁻ concentration (175–600 mg L⁻¹). At these points, the thickness of the unsaturated zone is limited, and the highest NO₃⁻ concentrations are recorded (<100 mg L⁻¹). Both of these factors suggest anthropogenic contamination of the aquifer. The remaining points in the Plioquaternary aquifer fall away from the FW–SW line of mixing (Fig. 5). This

indicates that the SO₄²⁻ and B are not exclusively associated with marine intrusion processes, and that other inputs of these ions must exist; specifically the leaching of evaporites and the presence of brines. The boreholes that tap carbonate rocks and have SO₄²⁻ concentrations of more than 100 mg L⁻¹ indicate a close relationship between B and SO₄²⁻: they are highly correlated ($r^2 = 0.75$). In addition, the line of best fit ($y = 0.001x - 0.014$) is very similar to the FW–SW line ($y = 0.001x + 0.022$) (Fig. 5). We have therefore interpreted the source of the SO₄²⁻ and B at these points to be directly associated with seawater intrusion. Two points within the carbonate aquifer with a high B content are separated from this trend, as a result of the presence of volcanic rocks.

Conclusions

An analysis of the distribution of boron in the groundwaters of the Aguadulce unit has allowed us to distinguish the processes that determined its source, namely its direct relationship with the lithology of the aquifer and anthropogenic influences, whether as a result of seawater intrusion or pollution from other sources. In the carbonate aquifers, the greatest concentrations are recorded at the points closest to the volcanic rocks (4 and 6 mg L⁻¹). At the other points, marine intrusion has been detected, and though the boron content is not elevated (<1 mg L⁻¹), the B/Cl ratio calculated is close to that of seawater, and so its origin has been related to this process. In the Plioquaternary aquifer, the situation is complex: this is a superficial aquifer with a very variable lithology which includes evaporites. Based on the boron content, it has been possible to distinguish between zones affected by anthropogenic pollution — with their elevated B content (2–14 mg L⁻¹), variable SO₄²⁻ (175–600 mg L⁻¹) and high NO₃⁻ (<100 mg L⁻¹) — and other zones where the source of the boron is related to the presence of evaporites. In the latter case, the boron content is greater than that on the FW–SW line of mixing obtained from a consideration of B, Cl⁻ and SO₄²⁻.

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