



Groundwater arsenic contamination and their variations on episode of drought: Ter River delta in Catalonia, Spain

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Abstract

The analysis and prospection using hydrogeochemical methodologies on arsenic (As) contamination episode in region of Spain (Girona, Cataluña) was investigated on the period (2000–2011), to analyze the mechanisms and characteristics of process on the solubilization of As in groundwater and effects of the episode of severe drought (2006–2007) on hydrogeochemical characteristics of the aquifer. The aquifer of study is a Mediterranean Delta; the geology of the zone has influence from sedimentary deposition of Ter River Basin and the prograde and draw back of the history coastline. The aquifer is an alluvial system where mean concentration of (As) is 30 µg/L in groundwater; parallel to this, the concentrations of elements iron (Fe) and manganese (Mn) exceed the guidelines of the World Health Organization (Guidelines for drinking-water quality electronic resource incorporating 1st and 2nd addenda, vol 1, Recommendations, 3rd edn, World Health Organization, Geneva, 2006) for drinking water. Range concentrations from other elements found in the groundwater were: Aluminum (Al, 4–716 µg/L), Barium (Ba, 48–603 µg/L), Cupper (Cu, 0.2–105.3 µg/L), Chrome (Cr, 1.1–47 µg/L) and Nickel (Ni, 1–51 µg/L). The arsenic solubilization trigger mechanism comes from desorption of oxide minerals and reduction related on neutral pH and reductive environment; the source of this kind of oxides is probably from marine sediments deposited in the process of delta's formation. During the climatic event of drought (2006–2007), the As concentration was responding to decline levels of the water volume on the aquifer, increasing their concentration and localized in a small area of the study zone, and these are effects of the reduction in groundwater flow on the aquifer.

Keywords Arsenic · Aquifer · Sedimentary basin · Drought

Introduction

The adverse effects in humans, caused by intake of arsenic (As) in diet elements, are a worldwide serious scope for the human health protection. Arsenic (As) is recognized as one of the most serious inorganic contaminants in drinking water in the world, particularly in regions where the concentration of arsenic in groundwater supplies is naturally high (Smedley and Kinniburgh 2002). Many areas around the world have problems with contamination related to human activities like mining and smelting activities, coal combustion,

tanning waste, pigment production, wood conservation, increased growth in feedlot-raised poultry and use of pesticides (Gomez et al. 2009; Pérez-Carrera and Fernández 2010). The contamination with As caused by human activities generally is localized and in almost all cases is not a health risk. Otherwise, natural pollution is present in regions with geological formations rich in arsenic, as plutonic rocks and sedimentary basins, used to occupy large geographical areas where water sources are compromised by the migration of arsenic aqueous forms to groundwater (Smedley and Kinniburgh 2002). This causes serious problems, for the water supply systems, and increases the population at risk (Anawar et al. 2006; Devi et al. 2010).

The process of (As) mobilization from aquifer substrate materials is regulated by hydrogeochemical process where the presence of oxidized and/or reduced mineral phases and the cofactors are associated with arsenic-rich solid phases. Arsenic-rich aquifers are characterized by elevated concentrations of dissolved iron, bicarbonate, ammonium

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ion and phosphate under reducing conditions and neutral pH (Anawar et al. 2006; Gomez et al. 2009; Hernandez-Garcia and Custodio 2004).

The species of (As) found in the environment are arsenic(III) and arsenic(V); the predominant species is determined by pH and reduction/oxidation potential. In many aquatic environments (As) species are in the form of oxyacid. Under oxidizing conditions (As), species are in the form of arsenic (V) or arsenate anions (AsO_4^{3-}), where (H_2AsO_4^-) is the most thermodynamically stable species at pH between 2 and 7 (Ryzhenko et al. 2009). In contrast, under reducing conditions the predominant species are As(III) or arsenite anions (AsO_3^{3-}), where neutral arsenite (H_3AsO_3) is thermodynamically the most stable species up to about pH 9 (Krainov et al. 2007; Ryzhenko et al. 2009). Arsenates, arsenate anions, and neutral arsenite constitute the main targets for field analysis, and they are actors in groundwater natural pollution (Pérez-Carrera and Fernández 2010).

The hydrogeochemical mechanisms for desorption of arsenic from the mineral pyrite are defined for the relationship of As with sulfate and iron. When the sulfide oxidation is the dominant process of the passing of As to its aqueous forms, a positive correlation can be found between the concentrations of (SO_4^{2-}) and (As) (Smedley and Kinniburgh 2002). In the sedimentary basins where the groundwater has a neutral pH and reducing environment associated with the presence of fresh organic matter, the geochemical environment is a determining factor for the solubility of arsenic (Selim et al. 2010). The basic trigger mechanism for the dissolution of As in porewaters environments of sedimentary basins, with neutral pH and reducing geochemical environments, can be associated to reduction-related dissolution of Fe and Mn oxides minerals, which results in the migration of As(III), Fe(II) and Mn(II) into the aqueous phase where they can reach high concentrations of these elements with a positive correlation between them (Smedley and Kinniburgh 2002), while the concentrations of (SO_4^{2-}) remains low (less than 1 mg/L). The high concentrations of phosphates, bicarbonates, silicates, and, possibly, organic matter may be favorable for As desorption (Anawar et al. 2006; Ryzhenko et al. 2009).

Of the regions of the world with groundwater arsenic problems, the worse emblematic episodes are localized on sedimentary basins (Smedley and Kinniburgh 2002). The iconic case of quaternary alluvial and deltaic sediment is associated with the Ganges–Brahmaputra–Meghna river system in Bangladesh, where more than 35 million people must drink groundwater containing (As) at concentrations greater than 50 $\mu\text{g/L}$ (Chakraborti et al. 2015). Other examples are the porewater from the Kalix River estuary of northern Sweden with concentrations in the range

1.3–166 $\mu\text{g/L}$ and porewater from clay sediments of Saskatchewan, Canada, where As was found at concentrations in the range 3.2–99 $\mu\text{g/L}$ (Wang and Mulligan 2005; Rahman et al. 2007).

The scope of this work is to analyze the origin and the trigger mechanism on solubilization of (As) in aquifer of Ter River delta. Associated with the identification of these processes, it is possible to see the effects of the drought period on the hydrogeochemical mechanisms and on the water quality of the aquifer studied.

Materials and methods

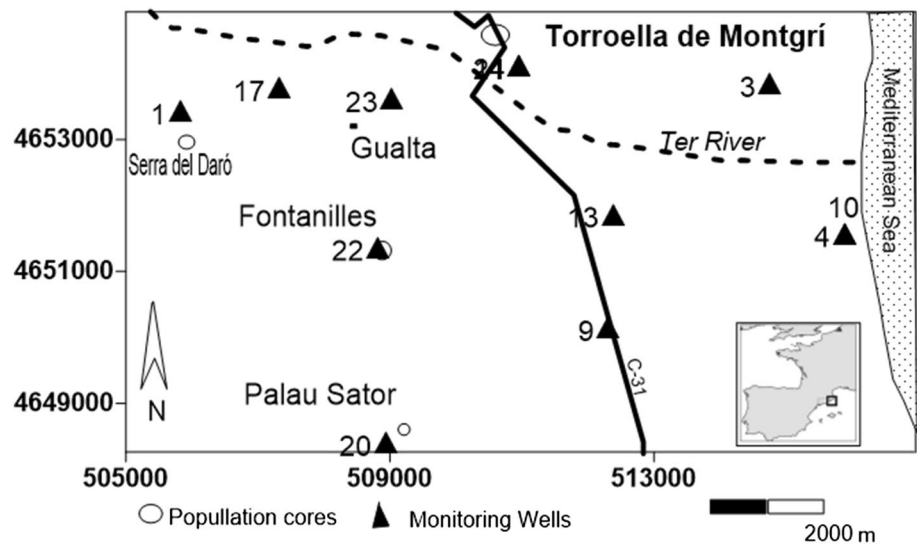
This research was realized analyzing information collected by the Agencia Catalana del Agua (ACA) through the monitoring program denominated “Register and Control of Quality of Waterbodies in Catalonia.” This is a 10-year (2000–2010) database with annual records of the concentration level of arsenic in the Ter River delta, and other aquifers of Catalonia, Spain, also includes the next water characterization parameters: electrical conductivity (EC), reduction–oxidation potential (REDOX), hydrogen potential (pH), temperature (T), major ions (SO_4^{2-} , Cl^- , HCO_3^{1-} , Ca^{2+} , Mg^{2+} , Na^+), and trace elements presences (As, Ba, Co, Cu, Cr, Sr, Fe). The sampling periodicity of the monitoring program was annual, generally effectuated in the period winter-spring on wells with different kind of uses basically a mix of municipal sources of water for human supply and private property wells used basically for agricultural camps of rice and fruit trees. The chemical analyses are made in certificated institutional laboratories.

To validate the information presented in the database, the Geochemistry, Petrology and Geological Prospecting Department of the Universitat de Barcelona (UB) in the year (2011), realized a sampling program, following the same protocol of ACA. The samples were sent to ACTLabs laboratories in Canada for its analysis on high-resolution inductively coupled plasma mass spectrometry (ICP-MS). The results reported by the ACA on 2011 were compared with the results of the UB sampling, and no significant difference was found between them.

Geographical and hydrogeochemical setting

The studied zone is localized in a rural activities territory of the state of Catalonia in the northeast of Spain (Fig. 1); it extends through the municipals demarcations of Gualta, Torroella de Mongrí, Palau Sator and Ullastret. The area is Ter River delta well known as Baix Emporda basin, an ecohydrology region formed for outfall delta in the Mediterranean coast. This delta has a quaternary sedimentary origin, which was developed as result of trailing sediments

Fig. 1 Baix Emporda, Ter River delta map showing the sampling sites



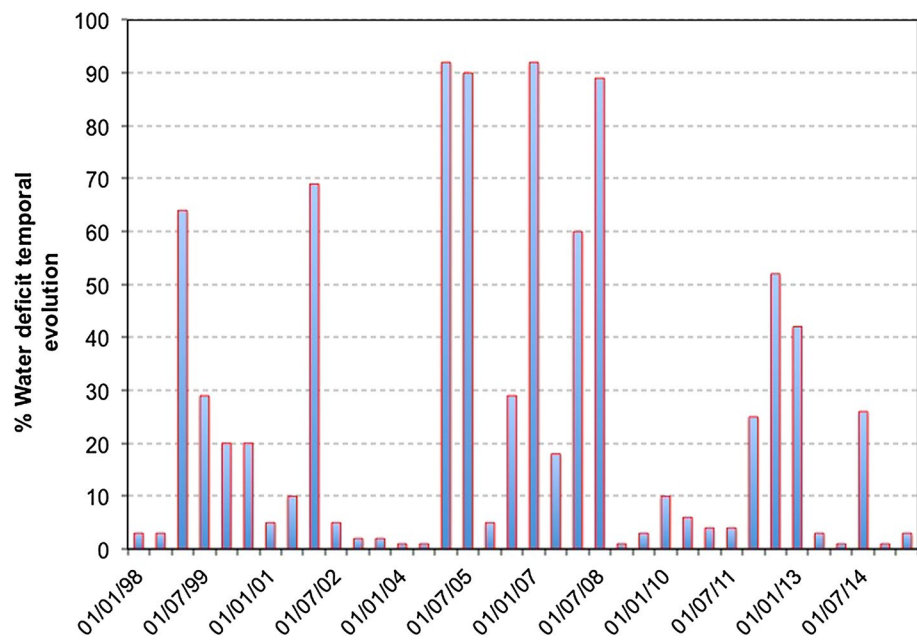
from Ter and Daró rivers through the historical process of interaction advance and retraction during the evolution of the coastline. This Delta is the end of a Ter River delta system with approximate 3010 km² area with a mean of rain of 740 mm per year. The groundwater delta system is formed for different permeability units layered sediments with mean 2772 Hm³ of capacity for water storage (Montaner and Subiranas 2009).

Drought episode

During first decade of 2000, the area of Catalonia suffers an episode of pluviometric drought, which affected an enormous region of Europe. In this climatic event, the water

sources of human supply technically were exhausted and provoke a serious restriction and extraordinary measures for all waters uses. In the period of 2005–2008, the water deficit was over 80% (Fig. 2) (Altava-Ortiz et al. 2017); due to consecutive years in rain deficit in April of the 2007, the authorities declare the emergency state for drought (Diario Oficial de la Generalitat de Catalunya, Decreto 257/2007). This situation continued through all 2007 without rain and the deficit grew almost to 90%. Until May of 2008, the situation was very difficult, the reserves of water a regional level was empty, and the emergency was alleviating in July month when it starts to rain and continues the next months. In January 2009 the emergency of drought was declared finished by the authorities (Agencia Catalana del Agua 2009).

Fig. 2 Water deficit temporal evolution period years 1998–2015 Source: Reproduced with permission from Altava-Ortiz et al. (2017)



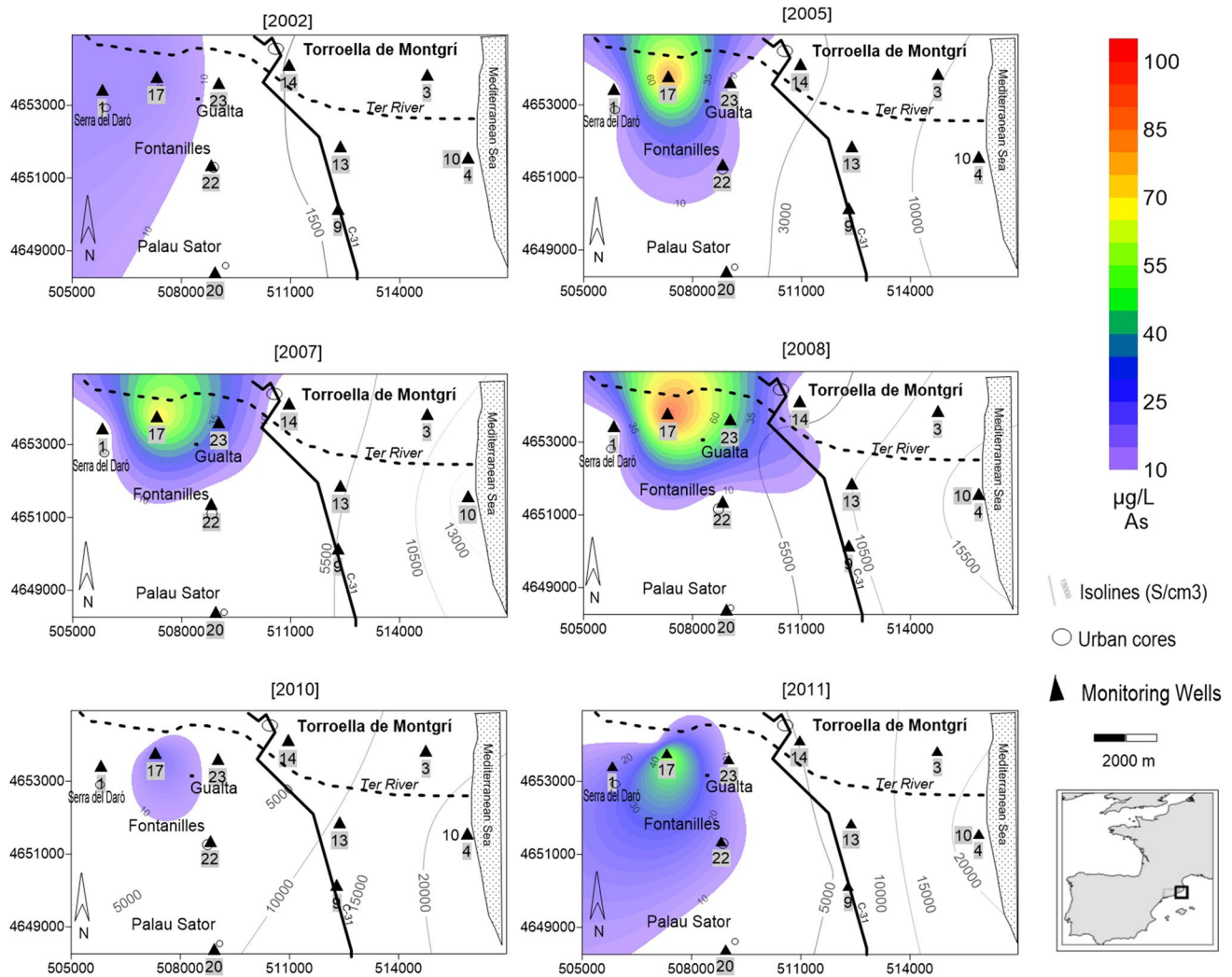


Fig. 3 Arsenic concentrations in periods

Results and discussion

On the study area, the concentration of As does not exceed the maximum limit recommended for the drinking water proposed for the WHO of $10 \mu\text{g/L}$ (WHO 2006), except for a small area of 6 km^2 , where the As concentration ranges from 20 to $90 \mu\text{g/L}$ (Fig. 3). Through the 10 years of observation, the concentration levels of As remain constant particularly in this zone. The general groundwater analysis (Table 1) presents metals which have sanitary important considerations, like Fe, Mn, Al, Ba and others.

The groundwater in this area is rich in calcium hydrogen carbonate (CaHCO_3) with evolution to (Cl–Na) (Fig. 4). The (Cl–Na) waters present a high EC values ($650\text{--}20,000 \mu\text{S/cm}$) because of marine intrusion, affected for the proximity of the line of coast (Fig. 3). In zones next to coast, the marine intrusion can affect the concentrations of As, by

dilution or ion exchange process that can reduce the As presence (Fig. 1) (Smedley and Kinniburgh 2002).

Trace elements

The presence of elements as (Fe), (Mn) and (Cu) (Table 1) is associated with As in sedimentary basins (Mor et al. 2006); they share hydrogeochemical mechanism of solubilization (Smedley and Kinniburgh 2002). These elements, particularly (Fe) and (Mn), are not correlated to the As concentration (Table 2), but its abundant presence is associated with high concentration of As in the area. Al and Ni showed high variation in area and time, and in some cases, they were above the WHO guidelines (2006). In this study was not found any relation between these elements and As as has been demonstrated by Puthiyasekar et al. (2010).

Table 1 Database of parameters from waters collected in the study area

Site	Year	E.C. µS/cm	pH	Eh mV	HCO ₃ ⁻ mg/L	SO ₄ ²⁻ mg/L	Cl ⁻ mg/L	Ca ²⁺ mg/L	Mg ²⁺ mg/L	Na ⁺ mg/L	Al µg/L	As µg/L	Ba µg/L	Co µg/L	Cu µg/L	Cr µg/L	Fe µg/L	Mn µg/L	Mo µg/L	Ni µg/L	Pb µg/L
3	2002	1178	7.8	N.N.	318	158	99	135	64	55	n.d.	n.d.	122	n.d.	n.d.	n.d.	178	941	n.d.	n.d.	n.d.
4	2002	12,468	N.N.	67	623	593	6803	407	3495	3495	n.d.	n.d.	234	3	14	n.d.	1105	2931	2	n.d.	n.d.
9	2002	1536	N.N.	176	467	230	155	185	61	98	62	n.d.	166	3	n.d.	n.d.	2091	945	n.d.	n.d.	13
13	2002	1666	N.N.	220	651	128	170	171	97	115	61	n.d.	82	3	n.d.	n.d.	4400	1211	n.d.	n.d.	n.d.
14	2002	1519	N.N.	216	587	167	178	180	114	63	72	n.d.	93	1	8	n.d.	1495	576	n.d.	n.d.	n.d.
17	2002	1032	N.N.	352	353	175	112	160	54	64	80	16	145	2	4	n.d.	2013	831	1	n.d.	n.d.
23	2005	873	N.N.	137	493	220	132	200	73	47	n.d.	6	239	n.d.	86	n.d.	1562	312	n.d.	43	n.d.
1	2005	878	N.N.	157	401	195	116	172	67	51	n.d.	n.d.	153	n.d.	n.d.	n.d.	831	408	n.d.	n.d.	n.d.
4	2005	18,250	N.N.	72	553	207	6611	180	4863	4199	n.d.	n.d.	207	n.d.	19	n.d.	11,250	4921	n.d.	n.d.	11
9	2005	1496	7.4	N.N.	534	134	109	141	91	112	n.d.	n.d.	123	n.d.	n.d.	n.d.	889	941	n.d.	n.d.	n.d.
13	2005	1194	7.3	N.N.	377	84	89.5	135	51	91	n.d.	n.d.	67	n.d.	n.d.	n.d.	223	1268	n.d.	n.d.	n.d.
14	2005	1387	7.4	N.N.	567	855	7243	420	4199	70	n.d.	n.d.	98	n.d.	n.d.	n.d.	361	703	n.d.	n.d.	n.d.
17	2005	768	N.N.	222	359	56	75.7	106	43	44	n.d.	88	106	n.d.	n.d.	n.d.	354	1172	n.d.	n.d.	n.d.
1	2007	932	7.2	N.N.	396	110	85	156	54	54	n.d.	n.d.	140	n.d.	11	n.d.	n.d.	n.d.	12	n.d.	n.d.
4	2007	14859	7	N.N.	593	957	6194	281	376	3888	n.d.	n.d.	130	n.d.	n.d.	n.d.	n.d.	3968	n.d.	n.d.	n.d.
9	2007	1040	7.3	N.N.	219	109	61	96	45	74	n.d.	n.d.	48	n.d.	n.d.	n.d.	262	888	n.d.	n.d.	n.d.
14	2007	N.N.	N.N.	N.N.	470	336	158.5	228	100	73	n.d.	n.d.	109	n.d.	n.d.	n.d.	1485	601	n.d.	n.d.	n.d.
17	2007	922	7.4	N.N.	440	98	76	138	541	54	n.d.	72	189	n.d.	n.d.	n.d.	1410	1485	n.d.	n.d.	n.d.
20	2007	793	7	N.N.	458	133	68.2	125	45	38	n.d.	n.d.	53	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
22	2007	998	7.7	N.N.	405	69	43.1	140	37	46	n.d.	n.d.	72	n.d.	10	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
1	2008	907	7.5	N.N.	744	106	7386	439	1000	50	40	n.d.	139	n.d.	24	n.d.	297	14	n.d.	1	1
10	2008	17,807	N.N.	N.N.	744	1033	7387	439	400	1007	716	n.d.	135	n.d.	n.d.	n.d.	n.d.	5926	n.d.	n.d.	n.d.
14	2008	N.N.	N.N.	N.N.	500	308	207.4	232	107	96	13	4	112	n.d.	8	n.d.	806	577	n.d.	3	n.d.
17	2008	900	7.4	N.N.	363	57	64.7	109	41	54	24	102	134	n.d.	2	n.d.	616	1302	2	1	n.d.
20	2008	827	7.1	N.N.	404	47	62.2	115	53	41	10	n.d.	55	n.d.	1	n.d.	n.d.	n.d.	n.d.	1	n.d.
22	2008	778	7.5	N.N.	370	91	79	141	50	42	17	n.d.	146	n.d.	6	47	n.d.	3	n.d.	n.d.	n.d.
23	2008	977	7.4	N.N.	455	224	457	248	101	135	19	21	603	n.d.	16	n.d.	10,400	791	3	1	2
1	2010	976	N.N.	N.N.	764	102	7867	459	1000	51	n.d.	n.d.	147	n.d.	21	n.d.	n.d.	n.d.	n.d.	1	n.d.
10	2010	24,100	N.N.	N.N.	504	567	7154	413	5395	3125	n.d.	2	157	n.d.	16	n.d.	n.d.	500	2	5	n.d.
14	2010	1476	N.N.	N.N.	374	89	83.7	145	50	91	n.d.	1	86	n.d.	2	n.d.	n.d.	267	n.d.	2	n.d.
17	2010	1051	N.N.	N.N.	442	226	138.9	190	85	52	n.d.	23	143	n.d.	3	n.d.	n.d.	500	1	5	n.d.
20	2010	807	7.1	N.N.	393	118	69.5	129	51	42	n.d.	n.d.	62	n.d.	23	n.d.	n.d.	8	n.d.	n.d.	n.d.
22	2010	602	N.N.	N.N.	403	67	45.8	139	41	42	n.d.	n.d.	135	n.d.	n.d.	n.d.	n.d.	249	n.d.	11	n.d.
23	2010	959	7.7	N.N.	380	95	1106	124	81	81	n.d.	3	231	n.d.	n.d.	n.d.	n.d.	16	n.d.	n.d.	n.d.
1	2011	640	8	40	380	88	85	146	18	52	n.d.	0	223	n.d.	1	n.d.	400	16	n.d.	2	0

Table 1 (continued)

Site	Year	E.C. $\mu\text{S/cm}$	pH	Eh mV	HCO ₃ ⁻ mg/L	SO ₄ ²⁻ mg/L	Cl ⁻ mg/L	Ca ²⁺ mg/L	Mg ²⁺ mg/L	Na ⁺ mg/L	Al $\mu\text{g/L}$	As $\mu\text{g/L}$	Ba $\mu\text{g/L}$	Co $\mu\text{g/L}$	Cu $\mu\text{g/L}$	Cr $\mu\text{g/L}$	Fe $\mu\text{g/L}$	Mn $\mu\text{g/L}$	Mo $\mu\text{g/L}$	Ni $\mu\text{g/L}$	Pb $\mu\text{g/L}$
4	2011	23,496	7	-93	763	1028	7865	460	4011	3567	n.d.	58	185	3	54	33	180	6960	4	16	1
17	2011	781	7	-100	391	117	65	128	19	n.d.	n.d.	n.d.	123	n.d.	3	3	60	1840	1	2	0
20	2011	627	7	127	402	69	44	142	15	43	4	1	55	n.d.	6	n.d.	210	n.d.	1	1	1
22	2011	772	7	84	340	67	63	138	21	40	544	1	151	n.d.	0	n.d.	n.d.	139	n.d.	2	0
23	2011	5590	8	154	475	245	145	200	32	91	n.d.	8	112	n.d.	n.d.	1	60	134	1	7	0

N.A. Not evaluated, n.d. not detected

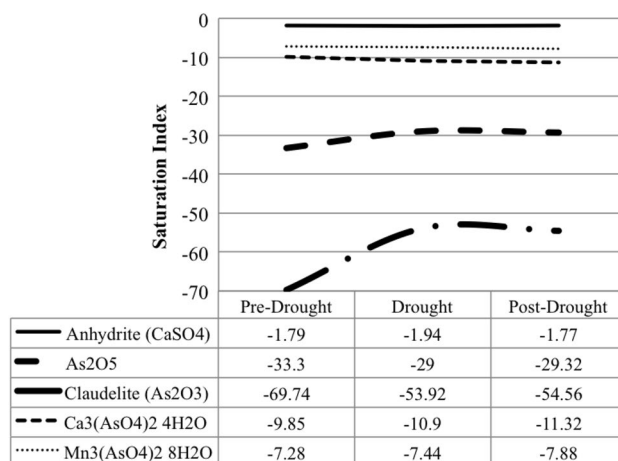


Fig. 4 Evolution of saturation index model of arsenic species in high concentration zones

Arsenic relationships and variations

The arsenic does not present statistic correlation with SO₄²⁻ or other majority ions, which indicates that the origin of As is not come from pyrite or arsenopyrites minerals sources associate with natural rich formations and mine activities (Table 2) (Smedley and Kinniburgh 2002). The low correlation values (zeros and negative) obtained between As and other ions in Table 2 show that it is not associated with these elements. This further supports the position that they exist in the form of oxides, which are solubilized in groundwater.

The neutral pH and the reductive conditions are in accordant situations, where desorption of oxide minerals and reduction-related dissolution of Fe and Mn oxides are the solubility mechanisms of (As) (Ryzhenko et al. 2009; Krainov et al. 2007). These conditions are common in aquifers systems like Baix Ter, with young quaternary sediments, mineral carbonate abundance rich in iron and manganese oxides, low gradient and semiarid environments (Smedley and Kinniburgh 2002; Krainov et al. 2007).

The speciation characterization (Fig. 4), made on a computer program for simulating chemical reactions and transport processes called PHREEQC (pH-Redox-Equilibrium-Concentrations) from the U.S. Geological Service, confirms explanation of mechanism desorption of oxide minerals, given that almost all elements (Fe, As, Mn, Cu and Pb) are in the oxide form. The As are in As(V) and As(III) species which are in negative saturation index, whereby solubilize in elemental forms, the same behavior happens for all other trace elements analyzed; oxides of (Fe, Mn, Cu and Pb) are solubilized in the groundwater with different intensities (Ryzhenko et al. 2009).

Another important clue to support the previous hypothesis is the geologic characteristics and morphology of Baix

Table 2 Correlations between most elements present in groundwater of study area

	HCO ₃ ⁻	SO ₄ ²⁻	Cl ⁻	Ca ²⁺	Mg ²⁺	Na ⁺	As	Ba	Cu	Fe	Mn
HCO ₃ ⁻	1										
SO ₄ ²⁻	0.386	1									
Cl ⁻	0.512	0.687	1								
Ca ²⁺	0.603	0.656	0.846	1							
Mg ²⁺	0.525	0.752	0.972	0.849	1						
Na ⁺	0.237	0.815	0.696	0.599	0.718	1					
As	-0.246	-0.191	-0.131	-0.223	-0.129	-0.112	1				
Ba	0.141	0.111	0.227	0.207	0.308	0.253	0.069	1			
Cu	0.175	0.035	0.078	0.015	0.108	0.046	-0.36	0.265	1		
Fe	0.147	0.425	0.363	0.256	0.408	0.489	-0.049	0.277	0.159	1	
Mn	0.454	0.72	0.842	0.742	0.834	0.682	0.03	0.067	-0.067	0.29	1

Emporda aquifer; the delta is an area where sea and land coexist, this aquifer is formed by a big number of deposit of different size materials; the origin of (As) could be a deposit of sediments rich in iron and manganese oxides from antic marine deposits (prehistoric wetlands) which now are part of the aquifer (Montaner and Subiranas 2009; Mondal et al. 2010). The increase in arsenic associates to site 17, because the well is drawing water from a specific deposit of sediments rich in arsenic oxides, as are explained for Montaner and Subiranas (2009) on the study of morphology of the Baix Emporda aquifer. This area situated around the site 17 represents the more affected area on the concentration of arsenic, in year 2008.

Drought effects in groundwater

The concentration of the majority ions was increased on the drought season declared by the authorities, basically caused by progression of the marine intrusion that affects more sampling wells (Fig. 3). The geochemistry milliequivalents analysis showed no variation between seasons (pre-drought, drought, post-drought). Basically, water facie (Ca-HCO₃) was constant in areas without marine intrusion affectation.

Elements (Al, As, Mn, Fe and Cr) increase their concentrations in period of drought, this behavior are common with drawdown volumes of water in the aquifer (Gonçalves et al. 2007). Meanwhile, Cu and Ni decrease their concentrations during the drought times.

The effect of climatic drought effect over the variations of (As) is evident in Fig. 3, the period before declaration of drought (years 2002) the concentrations of arsenic do not overpass the 15 µg/L, meanwhile in the drought period (2005–2008) the concentrations in a localized zone arrive to 80 µg/L in year 2005. The As concentrations are

continuously increasing reaching 90 µg/L a in the year 2008. This situation changes in the years 2010 and 2011 when the rain is back, and the effect of the drought is not present, reducing the As concentration to 10 µg/L in the studied zone.

That aquifer behavior is a clear evidence of drought effects in the piezometric evolution; when the rain scarcity is present, the groundwater flow and volume decrease, increasing the periods of residence parallel to the concentration of As and reducing the area of influence (Devi et al. 2010; Gonçalves et al. 2007).

Conclusions

The period of drought in the years 2005–2008 was accumulated an important humidity deficit in Baix Ter aquifer, causing an episode of natural contamination of groundwater, which was manifested by an increase in arsenic concentration in the groundwater extracted from a small area near Fontanillas town on the NE of Catalunya. The hydrogeochemical analysis showed the geological origin of the arsenic presence in the groundwater of the area, basically associated with dissolution of oxides of arsenic from sediments that form the geology of Baix Ter aquifer.

This episode was a side effect of drought, and it is evidence of the parallel affectations of the depletion of the levels of groundwater over the hydrogeochemistry of aquifers, and how this affects the potential use of this hydrological resources.

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