

Effect of saltwater intrusion due to over-exploitation and earthquakes on mineralization processes of spring waters over the Massif Bokkoya (central Rif, Morocco)

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ABSTRACT

The study focuses on the monitoring of physicochemical parameters and major chemical elements of six springs and wells during two seasons. Waters from the coastal karst aquifer Massif Bokkoya Al-Hoceima were analyzed for their chemical quality, and their state of anthropogenic and/or natural contamination was quantified. The waters are highly mineralized and marked by a chloride-sodium or sodium-sulfate facies mainly linked to a process of marine intrusion and dissolution of evaporitic minerals included in the on-site rocks. This mineralization of marine origin is partly due to the intensive exploitation of groundwater but also to landslides in relationship with earthquakes, that are known in the area, and which accelerate the phenomenon of saltwater intrusion. The salinity shows levels C3 and C4 in a Wilcox diagram and waters were qualified as very hard. The comparison with quality standards shows that most of the water is not suitable for consumption, especially because of high levels of electrical conductivity (EC), total dissolved solids (TDS), and Cl^- related to marine intrusion, together with the urban pollution factor which increases the content of NO_3^- .

Key words | contamination, hydrochemistry, limestone Bokkoya, marine intrusion, spring water

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INTRODUCTION

Coastal areas are defined as land areas whose environment, climate, and economy are strongly influenced by their proximity to the sea. They attract people both for their temperate climate and their quality of life, suitable for economic development. Over 60% of the world's population lives in these coastal areas (Tuthill & Calabrese 1981). However, these areas are at risk of water shortage, due to saltwater intrusion into groundwater reservoirs, often as a result of a strong exploitation of the underground resource.

Most residents of the Al Hoceima region prefer to consume water from springs and wells, which do not have the taste of chlorine used for treatment of tap water. However, the population ignores the quality of spring water and its potential deterioration by chemical and microbiological pollutants. The water springs and wells are easily subject to natural or anthropogenic contamination, especially in

karst areas where transfer velocities and infiltration are high. The intrusion of saltwater into aquifers is a phenomenon studied around the world, especially in the context of over-exploitation of coastal aquifers. In recent years, the process of intrusion in coastal karst aquifers has been better investigated by tracking changes in water quality. Many countries around the Mediterranean basin are affected by this risk, including Italy, Greece, Turkey, Croatia, and Spain, among others.

The central Rif is characterized by frequent landslides (Kortatsi 2007). Especially in 1994 and 2004, the region experienced two major earthquakes which resulted in a marked increase in spring water flow. The presence of faults caused by ground movements may also increase the salinity intrusion process. Earthquakes may also damage groundwater reservoirs, whose quality can be altered by a

pressure increase in confined aquifers that may result in a decreasing of dissolved oxygen changing redox condition causing the mobilization of some ions potentially harmful to health.

The limestone mountain chains play an important role in the water cycle of the Rif area. Their relatively large size, their position between low permeability series, the important local precipitations, their altitude, their karstification are all favorable to the large amounts of water storage which are available at the surface with only a limited delay. These conditions favor perennial streams, scarce in the Rif, and allow a water supply to rivers that cross the limestone in summer, and corresponding alluvial groundwater reservoirs (Hossien 2004).

Coastal heterogeneous aquifers and, in particular, coastal karst limestone aquifers are broadly divided into a porous or finely cracked matrix cut by open fractures or karst conduits created by tectonics and carbonates' dissolution (Arfib 2001). The hydrodynamic of these systems is dual, in conduits and in the host matrix. The water ideally flows in ducts, at high velocity, and is usually a non-Darcian flow. It is turbulent and occasionally exits at the surface or as an underwater spring (Mangin 1975). This duality of flows influences saline intrusion, which is disturbed by the flow in the conduits and can have a very heterogeneous spatial distribution (Arfib 2001). This work aims to characterize the geochemistry of localized springs in the Massif Bokkoya (Al-Hoceima) by analysis of major elements to evaluate the origin of the residual salinity.

ENVIRONMENTAL GEOLOGICAL AND HYDROGEOLOGICAL DATA

The study area is located in the Massif Bokkoya, in the eastern part of the Rif in northeastern Morocco (35.15–35.25° N; 3.85–4.00° W). The limestone mountain chain of the Rif is one of the major mountainous areas in Morocco, characterized by cracks' porosity developing important karstification. It covers an area of about 1,100 km², from the region of 'Sebta' (Ceuta, to the north) to the 'Al-Hoceima' (to the east). It is divided into three main units: the Haouz of Tetouan, Dorsal, and the limestone chain of Bokkoya (Dörfliger 2013). The climate is semi-arid and marked by

an alternation of a dry season (June to September) and a wet season (October to May) (Jones *et al.* 1999). Evaporation varies between 1,200 mm and 1,900 mm, lower in January and higher in July and August (30% of FTE). The Massif Bokkoya receives an average pluviometry of 300 mm/year. Recharge by infiltration was estimated at approximately 15 M.m³ on the chain of Bokkoya (Dörfliger 2013).

The Massif Bokkoya is a unit of the internal domain which outcrops in the north central part of the chain of the Rif (Figure 1). It consists of tectonic units forming the crystalline basement and covering Paleozoic and Mesozoic layers (Andrieux 1971; Mourier 1982). It is represented mainly by an internal limestone ridge, and an external limestone ridge that supports the form of tectonic klippe of Paleozoic land belonging to Sébtides and Ghomarides units (Barrocu 2003).

The limestone ridge borders the western and southern slopes of Ghomarides on the northern Rif. It constitutes a set of 'chips' to the massif Triassic-Liassic carbonate platform framework (El Hatimi *et al.* 1991). This series overlies condensed Jurassic-Cretaceous series, capped with a tertiary detrital material (Maychou 2009). Facing onto the Massif Bokkoya there is a narrow Plio-Quaternary corridor towards the subequatorial direction, possibly resulting from a filling of major synsedimentary faults with vertical displacement (Azzouz *et al.* 2002). As in all the southern Mediterranean rim, the Pliocene-Quaternary contains evaporite levels consisting mainly of gypsum.

The translation of the limestone-dolomite Triassic series of hedging Ghomarides over the Triassic dolomites of the dorsal external limestone is facilitated by the presence of red clay gypsiferous Triassic rocks (Barrocu 2003). These structural units consist of several tectonic slices of varying importance, stacked on each other. Their structure, characterized by essentially brittle deformation within the upper structural level, results from the superposition of several compressional and extensional phases that have followed from the Eocene to the present (Barrocu 2003).

Shales and evaporites (mainly gypsum) from Trias are intermingled with higher Cretaceous marl. These deposits are referred to locally as 'gypsum marl'. Locally these sediments have a breach character, also comprising blocks of metamorphic and plutonic rocks, referred to as 'polygenic breccia gypsum matrix'.

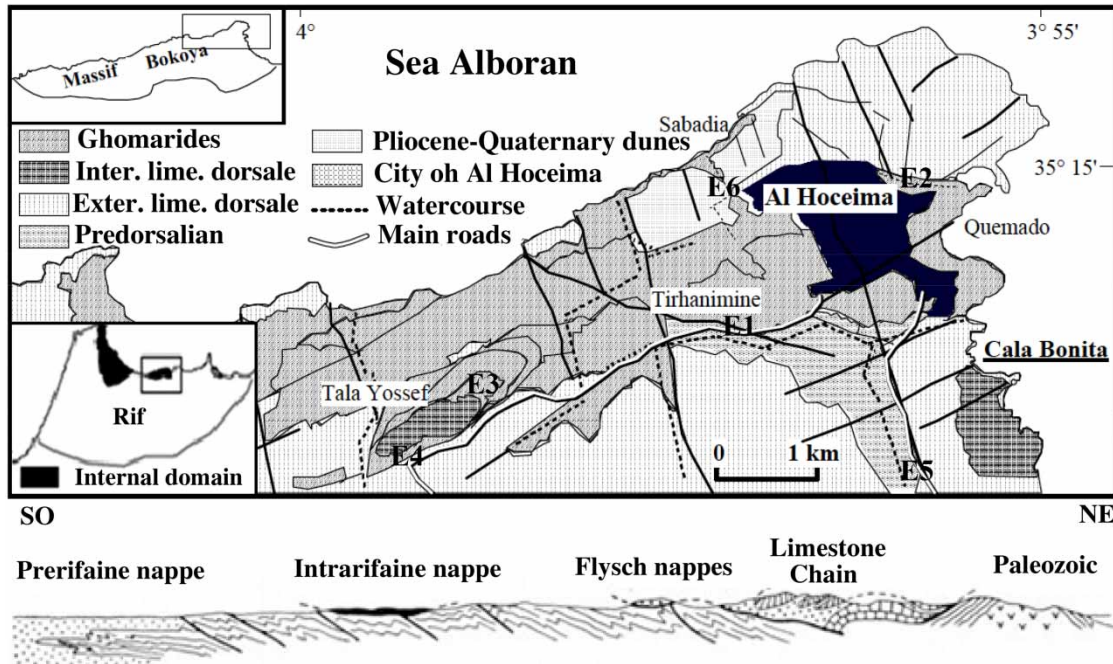


Figure 1 | Map and schematic geological section of the Rif (Lhénaff 1965). E1: watercourse, Dorsal limestone-Ghomarides; E2: city, land Paleozoic Ghomarides-dunes Pliocene-Quaternary; E3: Dorsal limestone; E4: Dorsal limestone-Ghomarides; E5: river, coastal pebble loam dunes Pliocene-quaternary; E6: lands Paleozoic Ghomarides.

The rainfall on the Massif Bokkoya seeps into the limestone formation or is quickly discharged at sea by torrential superficial flow.

During earthquakes, particularly in 1994, 2004, and 2016, abnormal flows of rivers in the Massif Bokkoya were observed, marked by a sharp increase in the momentary flow of springs. Carbonated, lapiazed materials may become highly fractured, increasing permeability, water infiltration, and ultimately, outflow. These may well be true groundwater perched above impermeable clay layers that line the shallow dip of contacts at the base of the different plies. The abundant fracturing in vertical and horizontal Dorsal limestone induces a rapid recharge on the surface and deep circulation materialized by many seeps (Azzouz et al. 2002).

MATERIALS AND METHODS

In 2015, two sampling campaigns were carried out over a year, during wet (3–15 February) and dry seasons (12–30 September). Four samples from each spring were taken at 3-day intervals during each campaign (values in Table 1

represent the mean value). E1, E2, E3, E5, E6 are managed springs, and E4 is a covered well with a depth of 15 m. The letter ‘H’ is used for wet season samples and the letter ‘S’ for the dry season. E6 was sampled only during the dry season.

Temperature, electrical conductivity, pH, and dissolved oxygen were measured in the field using a DZB-718 Portable Multi-Parameter Analyzer. Ions (HCO_3^-), (Cl^-), (Ca^{2+}), and (Mg^{2+}) were determined by the volumetric method. NO_3^- and SO_4^{2-} were determined by colorimetric assay using a spectrophotometer (UV/Vis). Measurements of potassium, sodium, and lithium were performed using a Sherwood Model 420 Dual Channel Flame Photometer.

RESULTS AND DISCUSSION

For all the samples, the ion balance was checked to be less than 10% (Table 1). Electrical conductivity (EC) was higher ($>1,800 \mu\text{S}/\text{cm}$) for E2, E3, E5, and E6. We can see that the conductivity varies between the wet and dry seasons, certainly consistently with the flow, the higher variation is observed for the E3 source (about 20%). The pH remains relatively constant between 7.1 and 7.9. Temperatures are mild in

Table 1 | Physicochemical analysis of spring waters in the Massif Bokkoya

Sample Season	E1		E2		E3		E4		E5		E6 Dry	Sea -	WHO (2011) -
	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry			
pH	7.53	7.15	7.58	7.34	7.4	7.28	7.85	7.58	7.44	7.24	7.34	7.70	6.5–8.5 ^a
T (°C)	18.0		18.0		18.0		18.0		18.0			18.0	-
EC (µs/cm)	1,366	1,187	2,430	2,130	2,230	2,690	594	602	1,802	1,993	2,110	56,000	1,250 ^b
TDS (mg/L)	1,288	989	2,231	2,104	2,528	3,110	444	531	1,828	1,958	1,878	-	1,000 ^a
Cl ⁻ (mg/L)	468.5	355.0	507.6	454.4	1,075.7	1,008.2	131.4	113.6	411.8	390.6	820.7	19,675	250 ^a
HCO ₃ ⁻ (mg/L)	280.6	183.0	320.3	280.6	231.8	256.2	183.0	219.6	335.5	366.0	248.4	152.0	-
SO ₄ ²⁻ (mg/L)	112.3	127.2	680.4	622.1	377.2	416.3	69.0	57.5	618.2	604.7	268.2	2,664.0	250 ^a
NO ₃ ⁻ (mg/L)	21.3	22.5	35.2	40.2	63.4	71.4	24.1	26.3	9.2	10.0	55.6	ND	50
Ca ²⁺ (mg/L)	72.1	67.3	216.3	190.6	240.3	213.1	43.3	41.7	195.4	190.6	68.9	414.0	^c
Mg ²⁺ (mg/L)	36.6	30.7	102.8	88.2	102.5	93.0	16.3	14.4	102.1	133.3	75.8	1,431	^c
Na ⁺ (mg/L)	273.6	227.2	348.0	358.0	576.0	564.0	128.0	124.8	273.6	266.4	460.0	11,216	200 ^a
K ⁺ (mg/L)	6.1	5.2	12.5	13.8	12.7	16.1	0.9	0.8	3.9	3.8	30.0	179.0	-

Only the guideline value is fixed for nitrate (50 mg/L) in consumable water (WHO 2011).

^aIndicative limit for taste.

^bIndicative limit issue from relationship between TDS and EC.

^cIndicative limit for hardness (Ca + Mg = 500 mg/L).

the rainy season and warmer in dry seasons thus well marked by seasonality. The measured total dissolved solids (TDS) show generally highly mineralized waters with the exception of E4. The annual mean temperature at Al Hoceima is 18.6 °C which corresponds to temperature values measured in springs and the well during the wet period. However, during the dry period, the temperature measured was between 21.0 and 28.5 °C, due to bad measurements in relationship with a re-equilibration between sampled water and atmosphere. For this reason, temperature values during the dry season are not shown in Table 1.

Geochemical analysis shows mainly the predominance of sodium and chloride except for E4, in which bicarbonates are higher quantitatively. Sulfates show an equal abundance with chlorides in E2 and E5 water in the dry season.

Nitrates are relatively high in E3 and E6, higher than the guideline value of 50 mg/L for nitrate in consumable water (WHO 2011). The higher concentration of nitrate is related to the locations of E3 and E6. Indeed, E3 (alt. 202 masl) is located in a lower part of an urban area, which does not have a sewage system. Local people pour their wastewater into individual septic tanks, and contamination may originate in the diffusion of elements from septic tanks into groundwater. E6 (alt. 17 masl) is close to the canal draining

wastewater from the city of Al Hoceima to a wastewater treatment plant. Leaks from the sewage canal probably cause significant levels of nitrates recorded in groundwater as well.

The results of physicochemical analyses of springs were compared with the different classifications according to the guidelines of the European Water Framework (JOUE 2009).

The dominant chemical profile corresponds to chloride-sodium (Figure 2). The sulfate-sodium and sodium-carbonate facies occur for E2 and E5 linked to mineralogy of Pliocene-Quaternary dunes.

Water classification according to mineralization depends on the content of the solid residue after evaporation of the water at 105 °C. We note that mineralization is low for E4 (E4 < 500 mg/L), moderate for E1 (900 < E1 < 1,500 mg/L) and higher for E2, E3, and E5 (>1,500 mg/L). TDS values for most samples exceed the recommended value by the WHO (TDS > 1,000 mg/L).

The low mineralization observed in E4 is probably due to its shallow depth (15 m) from where the vertical transfer is faster, the recharge taking place by direct infiltration.

Comparison of TDS between the two seasons shows a general increase or stable value in the dry season (E2, E3, E4, and E5). The difference is greater for springs E1 and

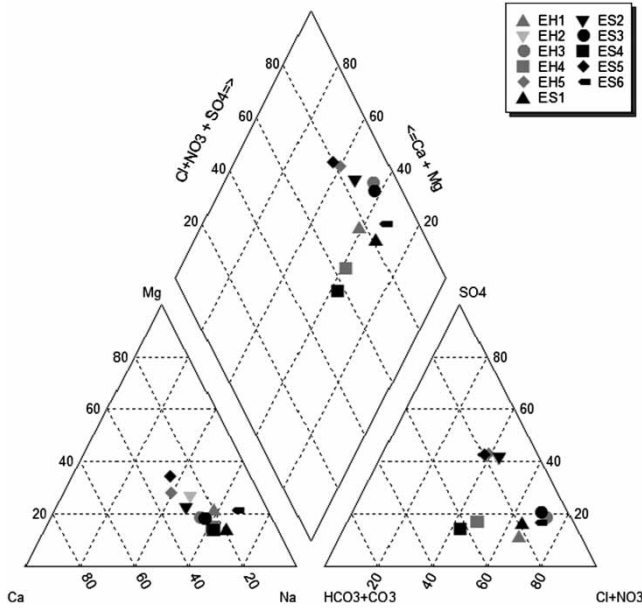


Figure 2 | Piper diagram: source of water at Bokkoya.

E3 and well E4 (20–25%) and weaker for springs E2 and E5 (5–7‰).

The different waters show high ions' variability except for HCO_3^- (Schöeller diagram, Figure 3). The heterogeneity and anisotropy characterizing the karst aquifers are highlighted in the strong geochemical variability of water. For

the well, E4, due to the shallow water level (15 m) and significant altitude (273 m), and the recharge by direct infiltration, the transit time is short and the salt content lower than other points.

Ion values are generally lower than 10 meq/L except for Na^+ and Cl^- which exceed 10 meq/L for most of the samples except E4.

The shape of the sample curves E2, E5, and E6, located at low altitude, imply the presence of sea water contamination in these springs, but with a high dilution factor.

The total water hardness (TH) expressed in mg of CaCO_3 was calculated. The majority of water in the Massif Bokkoya (E1, E2, E3, E5, and E6) is classified as very hard ($>300 \text{ mg/L CaCO}_3$). WHO recommends a maximum hardness of 500 mg/L CaCO_3 .

The relationship between $(\text{Ca}^{2+} + \text{Mg}^{2+})$ and $(\text{HCO}_3^- + \text{SO}_4^{2-})$ shows most of the points more or less aligned on the line $(\text{Ca}^{2+} + \text{Mg}^{2+} / \text{HCO}_3^- + \text{SO}_4^{2-} = 1)$, suggesting the levels of Ca^{2+} , Mg^{2+} , HCO_3^- , and SO_4^{2-} ions mainly come from the dissolution of calcite and gypsum, related to the lithology of formations in the region (Najib et al. 2016).

The value of absorbable sodium (SAR) provides information on the exchange of calcium ions base by sodium ions (Figure 4). This parameter is significant for water

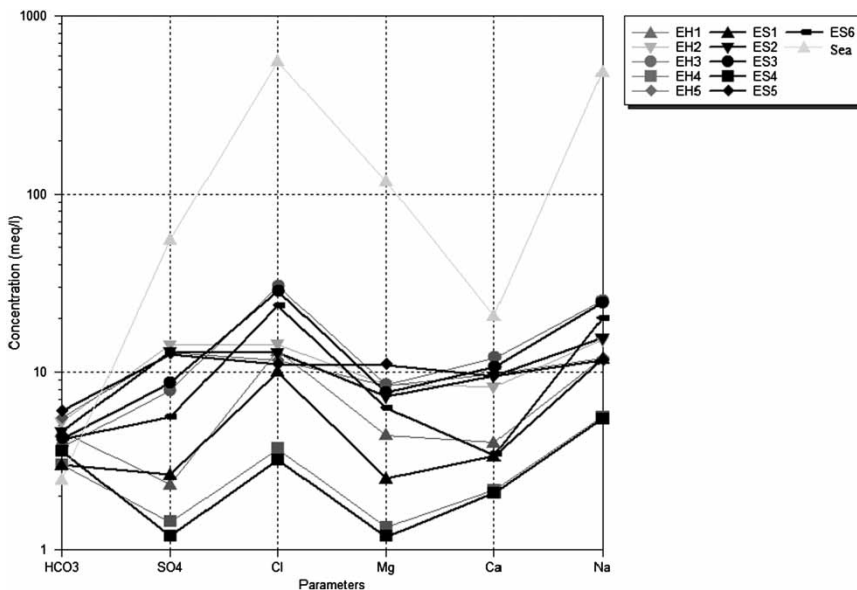


Figure 3 | Schöeller diagram.

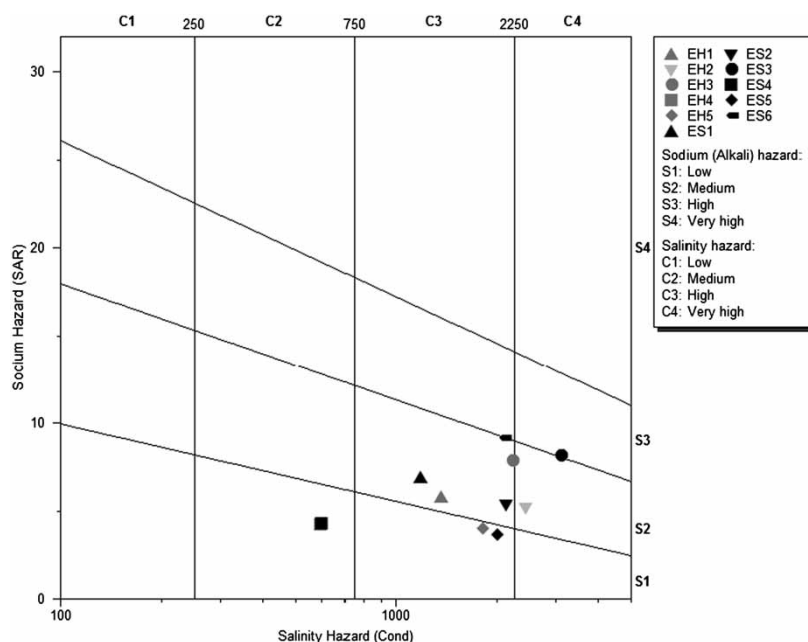


Figure 4 | Diagram of Wilcox log.

irrigation. Higher SAR values increase the hardness of the soil, reducing its permeability (Tijani 1994).

E4 water belongs to the C2S1 class, having very low salinity and a low risk of alkalizing. This means that water quality is consistently good with low mineral content. E1 and E5 waters belong to C3S1 and C3S2 classes; they are poor quality water with medium to high salinity and an alkalizing risk of low to medium. E3 and E2 waters belong to C3S2 and C4S2 classes, with poor quality and highly mineralized water, characterized by a salinity ranging from high to very high, and a medium alkalizing risk.

The difference in salinity between dry and wet seasons is low, except for E3, the most mineralized springs showing a high variability related to sea water contamination in the karst conduits linked to seasonality, the rains enhancing the flow of fresh water. This is consistent with other studies of coastal brackish springs in karst during floods which have identified three processes: the drain conduit karst, the dilution with fresh water, and the regulation of intrusion sea water into the pipe via the increase or decrease of load (Arfib 2006).

All Bokokoya samples are marked by relatively strong concentrations of sodium and chloride. Sodium levels in the water of E1, E2, E3, and E5 exceed 200 mg/L. Sodium

concentrations in drinking water are typically less than 20 mg/L. The human body needs about 120 mg/day of sodium chloride. A higher concentration of sodium in drinking water is suspected to act adversely on blood pressure, which may be elevated out of the norm (Tuthill & Calabrese 1981).

High levels of chlorides are common in coastal aquifers; they can be a marker of sea water intrusion (Ahmad & Bajahlan 2009). The chloride concentrations measured in the samples are very high; at the E3 spring they reach a value of 1,076 mg/L.

The molar ratio $[\text{Na}^+]/[\text{Cl}^-]$ in the case of saltwater intrusion is generally lower than the ratio of sea water (0.879), and presents values less than 0.86 in aquifers subject to saltwater intrusion (Rao & Rao 2009). Thus, the low ratios $[\text{Na}^+]/[\text{Cl}^-]$, combined with other geochemical parameters, can be an indicator of saltwater intrusion, even at relatively low concentrations of chloride in the early stages of salinization (Baker & Lamont 2001). The loss of sodium in sea water intrusion into the aquifer is mainly attributed to the cation exchange process by the balance carbonates. Elevated concentrations of Ca^{2+} and reduction of Na^+ levels were explained by the retention of Na^+ by exchange with Ca^{2+} and Mg^{2+} (Vengosh *et al.* 1991).

The $[\text{Na}^+]/[\text{Cl}^-]$ ratios of the E3 and E6 springs are less than 0.88, thus indicating a salinity mainly due to the intake of sea water. The values found for the springs E1, E2, and E5 and the well E4 are superior to 0.88. This difference can be interpreted by the coupling of sea water intrusion and exchange reactions with clay minerals where the Na^+ ions are released into the water replaced by Ca^{2+} and Mg^{2+} (Capaccioni *et al.* 2005).

To make assumptions about the mineralization origin of the springs, a first approach is to present the relationships between key major elements related to saline intrusion (Na^+ , Cl^-) and total solid residue (TDS) (Figure 5).

In general, the amount of NaCl in a coastal karstic aquifer does not exceed 2 g/L (Andrieux 1971). In our case, values are between 0.2 and 1.7 g/L with higher values for E3 and E6 (>1.2 g/L).

The weight contribution of Na^+ and Cl^- ions in TDS is between 40 and 70%. The highest values correspond to E3 and E6 while the lowest values correspond to E2 and E5. The importance of halite in TDS shows the salt contribution from marine origin in mineralization of water.

For the E6 spring, located at low altitude (17 m) and close to the coast, the proximity of the sea may induce high concentrations of Na^+ and Cl^- in the local recharge due to sea spray atmospheric deposition. Increased levels of NaCl can also be caused by overuse of the aquifers, lowering the groundwater level and locally promoting a mixture between fresh and sea water.

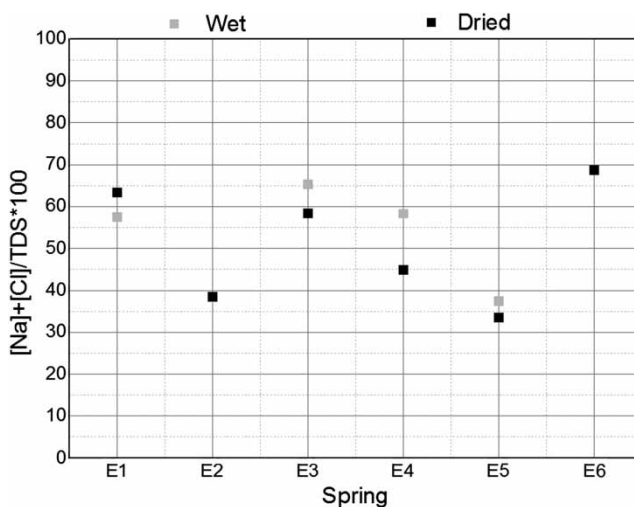


Figure 5 | $(\text{Na} + \text{Cl})$ compared to TDS.

CONCLUSION

The hydrochemical study of coastal groundwater in the heavily exploited karst of the Bokkoya area and its extension, showed the mineralization of water depends on several key factors: the dissolution of minerals from the geological formations in place, the ion exchange in water–clay interaction, and the contribution of sea water intrusion into the aquifer. The latter process can be accentuated by earthquakes to which the region is subject.

The dominant hydrochemical facies is a chloride-sodium type indicating the contribution of marine waters' contamination and evaporite dissolution, to freshwater salinization.

The different waters studied show a high hardness, a consequence of the dissolution of calcite and gypsum from geological formations defining the aquifer. For spring E3, high hardness is related primarily to saline intrusion.

However, the salinity level in the study area is heterogeneous. It is important for the E3 spring which belongs to the dorsal internal limestone; it is lower for springs located next to watercourses (E1 and E5). The characteristics of sample E3 are evidenced by the clear correlation between Cl^- and Na^+ . The molar ratio of $[\text{Na}^+]/[\text{Cl}^-]$ is lower than sea water for E3 and E6, while it is superior for the other waters.

The $\text{Na}^+\text{-Cl}^-$ in the solid dry residue TDS represents 40–70% of the total ions, showing the importance of the intake of sea salt in water mineralization, with variable mineralization depending on local conditions and the position of springs.

There was also a significant anthropogenic contribution regarding the E3 and E6 waters with high nitrate concentration highlighting the vulnerability of this geological karst formation to anthropogenic contamination from the surface.

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