



# Multidisciplinary approach to understand the salinization of fractured crystalline aquifers in semi-arid region

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**Abstract.** In semi-arid areas, groundwater (GW) represents the most reliable resource for water supply. In the semi-arid region of Ceará (Northeastern Brazil), GW of the crystalline fractured aquifers has a salinity higher than expected in this type of environment. This study implemented a multidisciplinary approach based on hydrodynamic (piezometric) and hydrogeochemical ( $^{18}\text{O}$ ,  $^2\text{H}$ ,  $^3\text{H}$ ,  $^{14}\text{C}$ , CFC,  $\text{SF}_6$ , Electrical Conductivity, major ions) data to understand the salinization processes. Results demonstrate that GW is generally recent and recharged by meteoric waters mainly through indirect infiltration from evaporated surface water. The study suggests that GW, originally bicarbonated, becomes progressively enriched in chloride due to the dissolution and leaching of salts, during the wet season, which come from meteoric inputs and have precipitated in the unsaturated zone and pond sediments during dryer periods.

**Keywords.** SDG6; Field observation

## 1 Introduction

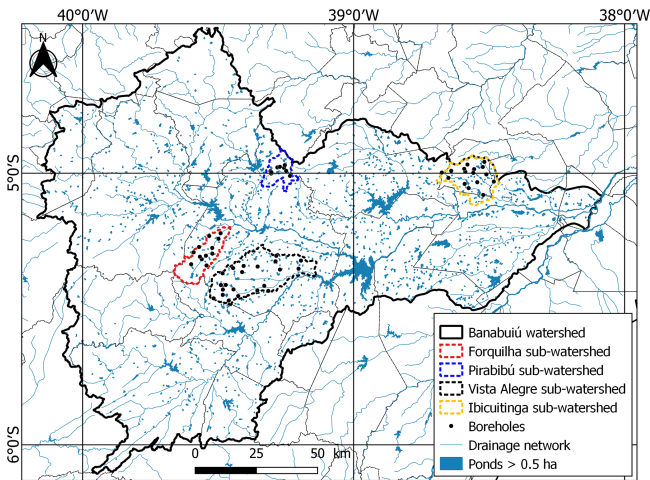
In crystalline aquifers, groundwater (GW) resources are generally scarce and may be subject to salinization, notably in semi-arid areas (Rabemanana et al., 2005; Jalali, 2007; Sreedevi et al., 2021). In semi-arid Northeastern Brazil (NEB), the processes at the origin of the high and heterogeneous GW salinity observed in the crystalline basement are still controversial. In Ceará (a state in semi-arid NEB), almost 75 % of the area consists of Precambrian basement rock (Santiago et al., 2001; Osorio, 2018). However, the hydrogeological functioning of the crystalline aquifers remains unclear despite the importance of this resource to cope with the lack of water in rural areas, especially during prolonged drought. Given the complexity of fractured medias, this study

used a multidisciplinary approach to understand the hydrogeological functioning and hydrochemical processes that lead to GW salinization through the implementation of a monitoring network of the electrical conductivity (EC) and the piezometric levels, a multi-tracer sampling ( $^{18}\text{O}$ ,  $^2\text{H}$ ,  $^3\text{H}$ ,  $^{14}\text{C}$ , CFC,  $\text{SF}_6$ ) and the analysis of major ions.

## 2 Study area

### 2.1 Geographic and climatic context

The study area is included in the Banabuiú watershed (19 647 km<sup>2</sup>), in the center of the Ceará at approximately 180 km of the oceanic coast. Investigations were carried out in four sub-basins of the Banabuiú watershed: Forquilha (214 km<sup>2</sup>), Pirabibú (127 km<sup>2</sup>), Vista Alegre (550 km<sup>2</sup>) and Ibicuitinga (286 km<sup>2</sup>), located between the latitudes 4°54'38" S and 5°29'08" S, and the longitudes 38°27'38" W



**Figure 1.** Map of the study area.

and 39°39'55" W (Fig. 1). These are rural areas of low human density, inserted in the Caatinga biome, whose socioeconomic development is mainly based on livestock and agriculture (MME, 1998).

The semi-arid climate of the study zone is characterized by water deficiency due to low precipitation levels ( $< 800 \text{ mm yr}^{-1}$ ), high evapotranspiration (PET) rates ( $> 1000 \text{ mm yr}^{-1}$ ), irregular rainfall distribution and recurrent drought periods (Burte, 2008; Martins and Júnior, 2017; Pontes Filho et al., 2020). The wet season, which extends from December to July, concentrates around 75 % of the precipitation over 4 months of the year (from February to May) in the study area. Moreover, these four months of rain are characterized by intense rainfall events (Costa and Herrera, 2016). The annual mean temperature is 27.2 °C, with monthly mean temperatures ranging between 25.7 and 28.5 °C. The monthly mean relative humidity varies from 41.6 % to 85.9 %, with an annual mean of 61.0 % (data from hydrometric INMET Station no. 82586, 1981–2010 period).

## 2.2 Hydrological context

In the crystalline zones of the study area, surface runoff is of Hortonian type due to the low thickness ( $< 1 \text{ m}$ ) and the low permeability of soils (Burte, 2008). In combination with the meteorological characteristics, all Ceará rivers are naturally intermittent (Santiago et al., 2001; Burte, 2008; Ribeiro Neto et al., 2022). The Banabuiú watershed is characterized by a large number of small and big dams, with more than 12 820 reservoirs mapped out by the FUNCEME (with an area greater than 0.5 ha). These reservoirs are subject to high evaporation losses and, consequently, to salinization and eutrophication problems (Santiago et al., 2001).

## 2.3 Geological and hydrogeological context

The Banabuiú watershed is characterized by the outcropping of the crystalline basement over 96.5 % of its surface. These rocks are represented by various types of gneisses and migmatites of Precambrian age, associated with plutonic and metaplutonic rocks of predominantly granitic composition (INESP, 2009). The remaining 3.5 % of the area are composed of Tertiary and Quaternary sediments (probably only Pleistocene according to Cavalcanti and Cavalcante, 2014) and colluvial-eluvial Quaternary covers. All of these Cenozoic sediments are interpreted as fluvial-lacustrine deposits (Da Silva et al., 2003).

The hydrogeological knowledge of the Ceará State is still very limited (Funceme, 2007). The presence of aquifers in the crystalline basement is related to the degree of alteration and fracturing of the rock, considering their very low primary porosity and permeability (Wyns, 2004; Lachassagne et al., 2021). In the study region, the median depth of water wells is 60 m and the level of the water table is generally around 10 m below the ground surface (Da Silva et al., 2007). Wells present low flow rates ( $1 \text{ to } 3 \text{ m}^3 \text{ h}^{-1}$ ). GW of the crystalline aquifers of Ceará may present very high EC values for this type of environment. No regional flow is observed according to Santiago et al. (2000), which means that the natural outlet of the crystalline aquifers is locally formed by the river or the alluvium (or a spring), at the scale of small sub-basins, unless evapotranspiration creates endorheic conditions.

## 3 Methodology and methods

Investigations were developed on two main axes. On one side, the study of the subterranean hydrodynamics thanks to the implementation of a piezometric monitoring network and the realization of a multi-tracers sampling ( $^{18}\text{O}$ ,  $^2\text{H}$ ,  $^3\text{H}$ ,  $^{14}\text{C}$ , CFC,  $\text{SF}_6$ ). On the other side, the study of the hydrochemical composition and evolution of GW thanks to the establishment of an EC monitoring network and the analysis of major ions. Studied borewells are distributed in the crystalline basement of the four sub-watersheds. All sampled borewells were pumped before sampling until stabilization of pH, temperature and EC parameters. Field parameters were measured in situ using an Aquaprobe® AP-700 multiparameter instrument.

### 3.1 Piezometric monitoring network

Monthly and hourly piezometric data were collected from March 2018 to December 2019 over a network of 56 boreholes, and from November 2016 to December 2019 in four of these boreholes, respectively.

### 3.2 Stable isotopes ( $^{18}\text{O}$ , $^2\text{H}$ ) analyses

The water stable isotopes ( $\delta^{18}\text{O}$ ,  $\delta^2\text{H}$ ) were measured on GW from 75 borewells (137 samples) sampled between August 2016 and August 2019. Analyses were performed in the water stable isotope analysis Laboratory (LAMA) of Hydrosciences Montpellier (France). Results are expressed in  $\delta\text{‰}$  vs V-SMOW (Vienna Standard Mean Ocean Water) as defined by Craig (1961). Analytical errors are  $\pm 0.08\text{‰}$  and  $\pm 0.8\text{‰}$  for  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$ , respectively.

### 3.3 Multi-tracer dating ( $^3\text{H}$ , $^{14}\text{C}$ , CFC, $\text{SF}_6$ ) analyses

GW samples were taken from 3 borewells in November 2009 and from 7 borewells in February 2018 for  $^{14}\text{C}$  and  $^3\text{H}$  analyses, and from 10 borewells in June 2019 for CFC and  $\text{SF}_6$  analyses.  $^{14}\text{C}$  and  $^3\text{H}$  analyses were carried out at the laboratory of Avignon University (France) and are respectively expressed as pMC (percent modern carbon) and TU (tritium unit). Analytical errors are about 0.5 pMC and 0.3 TU, respectively. CFC and  $\text{SF}_6$  analyses were performed at the Plateforme Condite Eau of Rennes University (France) and are within a precision of 3 % for CFC and 5 % for  $\text{SF}_6$ .

### 3.4 Hydrochemical analyses (EC, major ions)

An EC monitoring was carried out in conjunction with the monthly piezometric monitoring to evaluate the salinity dynamics of GW. Major ions sampling campaigns were conducted in December 2017 (dry season) and June 2018 (wet season) in 27 and 38 borewells, respectively. Major ions analyses were performed in the GECOQ laboratory of the CAGECE in Fortaleza (Brazil), in accordance with methodologies recommended by APHA (2012).

## 4 Results and discussions

### 4.1 Hydrodynamic characterization

#### 4.1.1 Piezometric monitoring

Piezometric monitoring evidenced that GW levels are relatively shallow, between 0.4 and 21.4 m deep. A contrasted temporal variation of piezometric levels was observed, with annual variations varying from 0.2 to 5.9 m depending on the boreholes. Seasonal recharge occurs during the rainy season, between February and May. Hourly piezometric monitoring showed a strong reactivity of GW levels ( $< 24$  h) to significant rainy events ( $> 50$ – $60$  mm), but a decrease of the level is almost never observed following the event. Instead, the levels gradually increase throughout the rainy season. Analysis of the variations in GW levels suggests that the seasonal recharge depends on the distribution and amounts of daily rainfall and is favoured by the storage of surface runoff in ponds.

#### 4.1.2 Stable isotopes monitoring ( $^{18}\text{O}$ , $^2\text{H}$ )

$\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values of GW samples showed a strong spatio-temporal variability and ranged between  $-4.04\text{‰}$  and  $+1.75\text{‰}$  and between  $-23.3\text{‰}$  and  $+8.0\text{‰}$ , respectively, with median values of  $-2.52 \pm 1.08\text{‰}$  for  $\delta^{18}\text{O}$  and  $-14.0 \pm 5.7\text{‰}$  for  $\delta^2\text{H}$ . GW presents low values of deuterium excess, with a median value of  $+6.1 \pm 3.6\text{‰}$ . The relationship between  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  of GW is expressed by Eq. (1):

$$\delta^2\text{H} = 5.09 \times \delta^{18}\text{O} - 1.22 \quad (r^2 = 0.9), \quad (1)$$

The slope of 5.09 of Eq. (1) is lower than the slope of the local meteoric water line (LMWL) defined in Kreis et al. (2020) and the global meteoric water line (GMWL), and clearly indicates that meteoric waters undergo significant evaporation before their infiltration (Chatton et al., 2016). Moreover, Eq. (1) is very similar to the linear regressions obtained by Matsui (1978) and by Stolf et al. (1977), cited in Matsui (1978) for surface waters in the NEB, which are respectively of  $\delta^2\text{H} = 4.6 \times \delta^{18}\text{O} - 0.5$  ( $r^2 = 0.97$ ) and  $\delta^2\text{H} = 4.75 \times \delta^{18}\text{O} - 1.3$ . Therefore, the strong similarity between the linear regressions of GW and surface water highlights that part of GW recharge occurs through a mixing process involving surface water marked by evaporation. This hypothesis is consistent with the numerous dams, lagoons and temporary drainage networks existing in the study area, and is supported by the GW level survey.

#### 4.1.3 Multi-tracer dating ( $^{14}\text{C}$ , $^3\text{H}$ , CFC, $\text{SF}_6$ )

The assessment of GW residence time through a multi-tracer approach showed that the apparent age of the GW varies from a few decades to hundreds of years. It also underlined the large contribution of post-1950s water in the recharge processes with  $^{14}\text{C}$  values generally higher than 100 pMC and measurable  $^3\text{H}$  values, but highlighted the complexity of flow path organization in the fractured crystalline system. Indeed, CFC and  $\text{SF}_6$  analyses, best interpreted by a binary mixing model (BMM), suggested the influence of water mixing between a recent pole (15 % to 85 % of the contributions) corresponding to a present-day recharge (fast vertical flows) and an older pole of longer transit time (interpreted as slow horizontal transfers). The BMM is based on the mixing of two piston flow water bodies. Contrary to the classical BMM which allows mixing between young (current CFC values) and old (CFC free) reservoirs, Condite Eau Plateform (Rennes University) tests the date allowing mixing with more or less young and more or less old waters. The result given is the best combination of mixing result (age and proportion) obtained for each tracers. The uncertainty in the model results depends on the adequacy between the results given by the different tracers and does not exceed 20 % and 10 years. Results of the multi-tracer dating are synthesized in Table 1.

**Table 1.** Results of the multi-tracer dating. FOR = Forquilha, PIR = Pirabibú, VA = Vista Alegre, IBI = Ibicuitinga.

Id. Samples	Sampling year	$^{14}\text{C}$ (pMC)	$^3\text{H}$ (UT)	CFC and SF <sub>6</sub> (Binary mixing model)	
				Recent pole (%)	Old pole (%)
PIR 44bis	2019	–	–	–	–
PIR 45	2019	–	–	85	15
FOR P7	2009	126.7	0.7	–	–
PIR P15	2009	124.4	1	–	–
VA Zuca	2009	119.5	0.9	–	–
PIR P16	2018	105.3	0.7	–	–
FOR 144	2018/9	103.6	0.6	65	35
VA 132	2018/9	110.1	≤ 0.7	45	55
PIR 52	2018/9	105.6	≤ 0.5	35	65
FOR 151	2018/9	98.3	≤ 0.7	50	50
IBI 89	2019	–	–	40	60
VA 123	2019	–	–	25	75
IBI 96	2018/9	73.2	≤ 0.4	20	80
IBI 152	2019	–	–	15	85
IBI 160	2018	91.8	≤ 0.3	–	–

## 4.2 Hydrogeochemical characterization

### 4.2.1 Electrical conductivity (EC) monitoring

The EC monitoring highlighted a strong spatial heterogeneity of the crystalline media, with EC values ranging between 886 and 19 310  $\mu\text{S cm}^{-1}$  (median value of 2850  $\mu\text{S cm}^{-1}$ ) and no salinity gradient. During the wet season, most boreholes show a temporary and fast increase of EC which suggests a leaching of salts stored in sediments, soil or in the unsaturated zone (USZ) during drier periods.

### 4.2.2 Major ions sampling

More than 75 % of the samples presented a mixed chloride or sodium chloride facies. However, the less salty waters (EC < 1600  $\mu\text{S cm}^{-1}$ ) presented a bicarbonated facies (< 10 % of the samples), whereas they tended towards a chloride facies with the increase in salinity. The chemical sampling before and after the rainy season showed a displacement of the points from the sodium pole towards the mixed pole, which suggests, in association with the observed increase in EC, a phenomenon of dissolution of calcium and/or magnesium salts (as calcite or dolomite). The strong correlation between EC and chloride ( $r^2 = 0.9$ ) or cations ( $0.7 < r^2 < 0.8$ ) and the absence of correlation between EC and bicarbonate, sulphate or nitrate ( $r^2 < 0.1$ ) indicates that the origin of high salinity is not related to the water/rock interaction or the anthropic pollution.

### 4.3 Origin of groundwater salinization

Among the processes that lead to salinization of GW, marine intrusions and the presence of marine evaporitic rocks are ruled out considering the distance from the study zone to

the oceanic coast and the absence of primary evaporitic terms in the Precambrian crystalline basement of the region. Moreover, the hypothesis of a past marine transgression is incompatible with the measured residence time of water. Mineral hydrolysis of crystalline rocks may explain part of the water mineralization, but not the predominance of chloride ion or the high salinization of GW. Considering that the chlorides and the GW salinity are not related to anthropic pollution, it results that the chloride source is external to the aquifer and derives from meteoric inputs (in the form of wet or dry depositions). Furthermore, the chemical study suggests that the GW, originally bicarbonated, becomes progressively enriched in chloride due to the dissolution and leaching of salts that were precipitated in the USZ and pond sediments during the dry season owing to the strong evapotranspiration rates, as observed in Madagascar (Rabemanana et al., 2005) or India (Sreedevi et al., 2021), for example. The precipitation of salts on the surface is favoured by alternating dry and wet periods (Sreedevi et al., 2021). The hypothesis of leaching of salts is supported by the observation of evaporite minerals (calcite, dolomite, gypsum, halite and probably bischofite) on the banks and sediments of the dams in the study area (Laraque, 1991; Araújo, 2017) and the presence of evaporated water in the aquifers.

## 5 Conclusions

This multidisciplinary study used hydrogeochemical and isotopic approaches, associated with piezometric data, to understand the hydrogeological functioning and the origin of the salinity of a complex crystalline system. Generally, the contrasted temporal and spatial variations of piezometric levels or hydrochemical composition of GW highlighted the strong heterogeneity and compartmentalization of the crystalline

aquifers, as well as the influence of local processes. The combination of results allows to identify that GW recharge occurs through both direct and indirect infiltration, with a significant proportion of evaporated surface water. The GW residence time is relatively short (significant supply of post-1950s water) and the flow paths organization is mainly composed of rapid vertical flows. In these aquifers, the salinity is interpreted as originating from rainfall. The evapo(transpi)ration of small rainfall events or surface waters during the dry periods favors the concentration of atmospheric salts on the surface. This study suggests that the GW becomes progressively salinized due to the dissolution and leaching of evapoconcentrated salts precipitated in the unsaturated zone and pond sediments, given the high evapotranspiration in the partially endorheic context.

**Data availability.** The research data presented in this study can be found in Kreis (2021), accessible at: <https://www.theses.fr/2021MONTG031>.

**Author contributions.** MBK carried out the investigations and interpretations of this study under the scientific supervision of JDT, CL, PL and ESPRM. JDPB carried out the 2009 sampling campaign for the measurement of  $^{14}\text{C}$  and  $^3\text{H}$ . NP and VVA performed respectively the stable isotope and the CFC/SF<sub>6</sub> analyses. MBK prepared the manuscript with contributions from all co-authors.

**Competing interests.** The contact author has declared that none of the authors has any competing interests.

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