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Isotopic characterisation and dating of groundwater recharge mechanisms in crystalline fractured aquifers: example of the semi-arid Banabuiú watershed (Brazil)

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ABSTRACT

Sustainable groundwater management implies a good knowledge of recharge processes, especially in areas with water deficit, like the semi-arid region of Banabuiú watershed (Ceará State, Northeast of Brazil). In this zone, phreatic aquifers consist of Precambrian crystalline fractured reservoirs characterised by a high spatial anisotropy, both in terms of hydrodynamics and water quality. This study implemented a multi-tracer approach (^{18}O, ^{2}H, ^{14}C, ^{3}H, CFC, SF_6), combined with hydrodynamic data (i.e. groundwater levels) to identify the groundwater recharge origin and the recharge mechanisms, and to estimate the groundwater residence time. At the basin scale, hydrodynamic data and local observations indicated the high reactivity of aquifers to precipitation and suggested that infiltration processes occur mostly through preferential infiltration zones. Stable isotope data showed a major contribution of evaporated surface water in the recharge process from many artificial or natural ponds. Groundwater residence time determination highlighted the spatio-temporal heterogeneity of flow path organisation within aquifers, with variable contributions between fast vertical flow (present-day end-member; 15–85 %) and a slower horizontal flow (old end-member <1960), underlining the vulnerability of aquifers to present-day environmental stress or pollution.

Keywords: Age dating; Brazil; carbon-14; crystalline aquifers; groundwater recharge; hydrogen-2; hydrogen-3; isotope hydrology; multi-tracer approach; oxygen-18; semi-arid; stable isotopes

1. Introduction

Given the scarcity of surface water in arid or semi-arid regions, groundwater (GW) constitutes generally the only reliable water resource for water supply. In fractured crystalline aquifers, GW resources strongly depend on present-day recharge capability [1]. Therefore, the identification of the GW recharge mechanisms is an essential knowledge to understand hydro-geosystems functioning and to evaluate the sustainability of these water resources [2]. Compared to porous media, the flow and transport processes in fractured media are complex, because GW flow is localised within the discontinuities of the rock (i.e.

fissures and fractures network) and is mainly controlled by the geometrical properties of the fracture network [3]. Due to this high heterogeneity and anisotropy of the media, the characterisation of GW flows in fractured hard rocks is generally quite challenging [3,4]. In crystalline basement areas, hydrogeological studies are of primary importance given the difficulties to quantify the GW recharge using conventional water balance and/or hydraulic methods. Previous research highlighted that isotopic methods are valuable tools for the characterisation of recharge processes and flow mechanisms in fractured hard rocks [2,4,5]. Indeed, the analysis of stable isotope contents on the different compartments of the water cycle (i.e. precipitation, surface water and GW), combined with a GW residence time determination, constitutes a powerful integrative approach to understand the GW origin and flow path organisation within aquifers, and to identify the relationship between surface water and GW [2,4–8].

In the Ceará State (Brazil), almost 75 % of the area consists of Precambrian basement rock that outcrops under a semi-arid climate. Despite the little aquifer potential of the crystalline basement, the GW contained in these aquifers enabled many cities to overcome the last prolonged drought (i.e. 2012–2016) thanks to the supplementation of the GW into the water distribution system of municipal networks or into strategic public water fountains. However, the global hydrogeological functioning of the Precambrian basement aquifers remains to this day very insufficiently understood knowing the importance of this resource to cope with the lack of water, in a region where pressures on water resources continuously increase. This study applied a multi-tracer approach (^{18}O, ^{2}H, ^{14}C, ^{3}H, CFC, SF_6), combined with hydrodynamic data (i.e. GW levels), in order to (1) identify the GW recharge origin, (2) understand the recharge mechanisms of the Precambrian basement aquifers, and (3) estimate the GW residence time. This multi-tracer approach is the first such attempt in the semi-arid region of Ceará and will contribute to a better knowledge and management of this resource.

2. Study area

2.1. Geographic, climatic and environmental context

The study region is located in the Ceará State, in the northeast of Brazil, between the latitudes 4°54′38″ and 5°29′08″S and the longitudes 38°27′38″and 39°39′55″W (municipalities of Quixeramobim and Ibicuitinga; Figure 1). The study area includes four sub-basins which are all part of the Banabuiú watershed, whose eponymous river drains an area of 19,647 km^2 in a west-to-east direction.

The climate is semi-arid and characterised by a strong spatio-temporal heterogeneity of precipitation, high evaporation indices, and recurring droughts. The rainy season extends from December to July, with more than 70 % of the rains concentrated in the period from February through May. These four rainy months, locally known as Quadra-chuvosa, are related to the Inter-Tropical Convergence Zone (ITCZ) influence, where the northeast and southeast trades flow together. The ITCZ is characterised by strong upward motion and heavy rainfall in the north of the northeast of Brazil [9]. The mean annual rainfall of the Banabuiú watershed, calculated from the Thiessen polygon method for the 1981–2010 period, is about 709 mm (ranging from 257 to 1077 mm in the same period), while the mean annual potential evapotranspiration is 1831 mm, which leads to a high water deficit in the area (>1000 mm). The 1981–2010 period is currently taken as official national climatology period used as reference for meteorological variables’ statistics computation. The 2012–2016 period corresponded to the most severe 5-year drought recorded in the systematic record. If considering the 2011–2019 period, the mean annual rainfall of the region would be about 580 mm (INMET Station n°82586 of Quixeramobim). Monthly mean temperatures range between 25.7 and 28.5°C, with an annual mean of 27.2°C, while the relative humidity presents monthly mean values from 41.6 (July) to 85.9 % (February), with an annual average of 61.0 % (data from hydrometric INMET Station n°82586 of Quixeramobim, 1981–2010 period).

One of the characteristics of the study area is that all the rivers of the Ceará State are naturally intermittent. The recession time of rivers is extremely short [10]. The drainage network is dendritic, the density and orientation of the drainage being influenced by topography, lithology and tectonic structures. In the crystalline zones of the study, the flows are essentially Hortonian because of the low thickness and the low permeability of the soils. However, only intense rains cause significant flows and favour the filling of superficial reservoirs [11]. The recurrence of drought periods and the intermittency of watercourses has pushed public policies to support dam construction projects, in order to guarantee water supply during periods of water stress [10,11]. The Banabuiú watershed is characterised by its large number of artificial or natural ponds (Figure 1), with more than 12820 reservoirs mapped out by the FUNCEME which have an area greater than 0.5 ha, and 1415 dams with an area greater than 5 ha [12]. The water storage in these dams is subject to high evaporation losses and, consequently, to salinisation problems [10]. Given the climatic pressure, many efforts have been made to build boreholes in order to ensure the water supply for rural populations during prolonged low-flow periods.

The study area corresponds to four sub-basins of the Banabuiú watershed that have the particularity of having already been studied (historical data; [11]). These are the sub-basins of Forquilha (FOR; 214 km^2), Pirabibú (PIR; 127 km^2), Vista Alegre (VA; 550 km^2), located in the municipality of Quixeramobim, and the Ibicuitinga sub-basin (IBI; 286 km^2), located between the municipalities of Ibicuitinga and Morada Nova (Figure 1). In terms of geomorphology, the altitudes of the Banabuiú watershed range between 1105 and 21 masl (maximum and minimum values outside the study area), the highest relief being observed in the western part of the basin. The different sub-basins present an undulating to slightly undulating relief. Their characteristics are summarised in the Supplemental material, Table SM1.

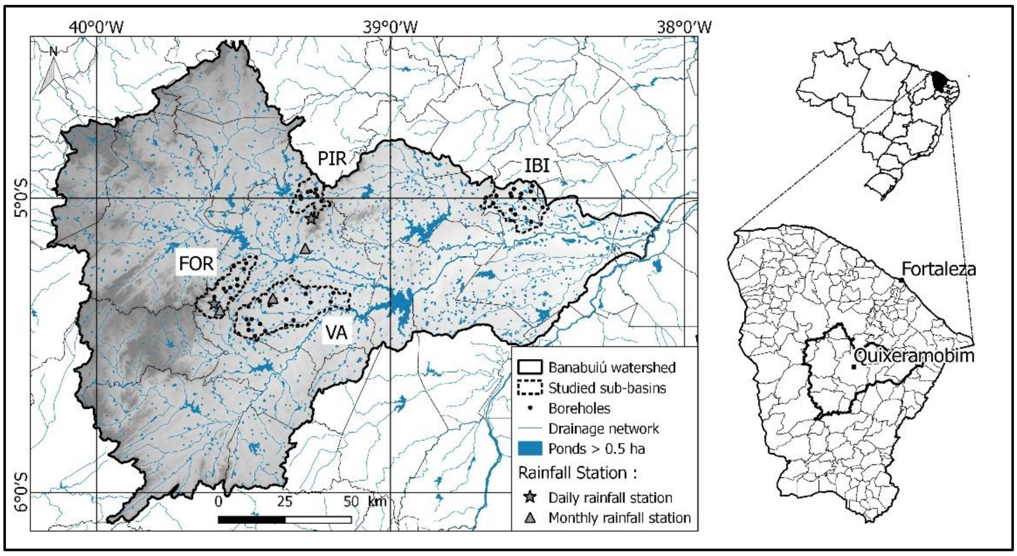

Figure 1. Localisation of the study area.

2.2. Geological and hydrogeological context

Almost 96.5 % of the area of the Banabuiú watershed is characterised by the outcropping of the Precambrian basement, composed of magmatic and metamorphic rocks of the gneiss type, migmatites, as well as plutonic and metaplutonic rocks of predominantly granitic composition [12]. The different lithostratigrafic units are oriented NE–SW in accordance with the main fracturing (Figure 2). The basement is covered on the remaining 3.5 % by tertio-quaternary covers corresponding to deposits of colluvio-alluvial formations (sandy clay or clayey sand sediments). The Ibicuitinga watershed, which is the flatter basin, is the only watershed that presents these sedimentary formations.

In the crystalline basement, the presence of an aquifer is related to the degree of weathering and fracturing of the rock, considering that the primary porosity of the crystalline rocks is almost zero. Due to the semi-arid climate, the alteration mantle of the rock is generally thin or inexistent [13], which implies that most of the water storage occurs in the network of fissures and fractures [14]. The absence of the alteration mantle may allow a direct connection between the surface drainage system and the fracture network.

Typically, crystalline basements have little aquifer potential because of their low permeability. In the study area, water wells generally have low-flow rates (≈ 1–3 m^3/h), and have an average depth of 57 m [15]. According to Santiago et al. [16], there is no regional subterranean flow within the Cearense crystalline aquifer. In addition to the quantitative problem, the water contained in crystalline aquifers of the semi-arid region of Ceará is subject to significant problems of salinisation. Based on data from 1991 boreholes implanted in the crystalline basement, Burte [11] highlighted wide ranges of electrical conductivity (EC) with values higher than one would expect in this type of environment (median EC value of 2375 µS/cm). The strong heterogeneity and anisotropy that can be encountered in the Precambrian basement aquifer system imply that boreholes exploit isolated fractures, which explains the great differences in chemical composition, static levels or flows that can be observed between two neighbouring boreholes.

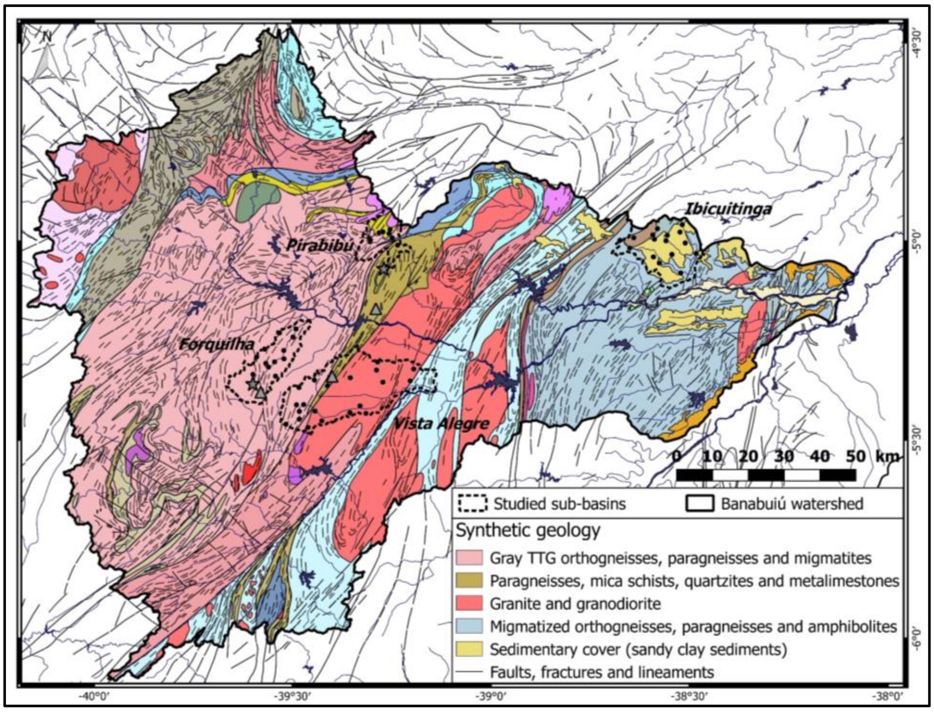

Figure 2. Synthetic geological map of Banabuiú watershed.

3. Materials and methods

3.1. Groundwater survey

A monthly piezometric and physicochemical monitoring (pH, temperature, EC) has been set up since April 2018 to observe the hydrodynamic response of the aquifer to precipitation, over a network of 56 boreholes implanted in the crystalline aquifer and distributed between the four sub-watersheds. In addition, 77 measurements sporadically conducted on this network between August 2016 and February 2018 were added to the database. The monthly GW level monitoring was supplemented by high-resolution piezometric monitoring (hourly frequency) from three pressure sensors. Boreholes equipped with a pump had level measurements carried out before pumping or under conditions close to the static level. Monthly physicochemical parameters were measured in situ, after a 15 min pumping.

3.2. Groundwater and surface water sampling

Stable isotope ($\delta^{18}O$, $\delta^{2}H$) analyses of water were developed over different hydrological periods (end of the wet and dry seasons) between August 2016 and June 2018 on 59 boreholes (118 samples) and 3 surface water sampling sites. Moreover, historical stable isotope analyses, taken quarterly from the study area between August 2009 and December 2012, were added to this recent database (12 boreholes and 10 dams, 62 samples each). For the GW residence time determination, GW samples were collected in November 2009 (3 samples) and February 2018 (7 samples) for ^{14}C and ^{3}H analyses, and in June 2019 (10 samples) for CFC and SF_6 analyses. Each GW sample was collected after a 30 min pumping or after the stabilisation of the physicochemical parameters (pH, temperature, EC), which ensured a good representation of the aquifer.

3.3. Rainwater sampling

The study of the isotopic composition of precipitation is a powerful tool to characterise the input signal of GW [17], as shallow GW resources generally reflect the long-term mean isotopic composition of precipitation. Thus, three experimental rainfall stations were operated between January 2011 and July 2019 in the municipality of Quixeramobim in order to implement a monthly monitoring of the isotopic composition of rainwater. Due to the impossibility of burying the collection system, the water collector was stored inside an isothermal box lined with aluminium foil, in order to minimise the effects of evaporation during the monthly storage. For technical and economic reasons, sampling was carried out around the 1st of the month (±48 h) between January and July, period in which 95 % of the annual precipitation is concentrated. Because of the high risk of evaporation that may occur inside the water collector during the monthly storage, two existing conventional rainfall stations of Quixeramobim were adapted in 2018 to implement a daily monitoring of the rainwater isotopic signal (Radar and Riacho Verde stations). Water collectors were adapted with the same technique used for monthly monitoring. The daily rainfall sampling is being carried out every day of the year around 7am, since March 23, 2018. The descriptions of the five rainfall stations are summarised in the Supplemental material, Table SM2.

3.4. Isotope and dating analyses

All rainwater, surface water and GW samples for stable isotope analyses (^{18}O, ^{2}H) were packaged in transparent plastic bottles of 20 ml, hermetically closed by an inner cap and a plastic cap. After sampling, samples were stored in a dark refrigerated container before being transported to the laboratory in France. Stable isotope analyses were performed in Montpellier (France) at the water stable isotope analysis laboratory (LAMA) of Hydrosciences Montpellier (HSM). Results are expressed in δ ‰ vs V-SMOW as defined by Craig [18]. The analytical error is ±0.08 ‰ for $\delta^{18}O$ and ±0.8 ‰ for $\delta^{2}H$.

The GW ^{14}C and ^{3}H analyses were performed at the laboratory of the University of Avignon (France), while CFC and SF_6 analyses were carried out at the Geosciences laboratory of the University of Rennes (France). ^{14}C activities are expressed as pMC (percent modern carbon), with pMC = 100 % corresponding to post-1950s infiltrated waters. ^{3}H activities are expressed in tritium units (TU). Analytical errors are respectively about 0.5 pMC and 0.3 TU.

4. Results and discussion

4.1 Isotopic characterisation of precipitation

The isotope monitoring of rainwater conducted since 2011 aims at defining the isotope input signal of GW resources. During the 2011–2019 period, the annual mean precipitation was of 580 mm. However, it appears that the years 2012–2016 were particularly affected by the drought, with a decrease of the annual precipitation of about 40 % compared to the 1981–2010 period ($P_{\text{annual mean 2012–2016}}$ = 434 mm). Isotope analyses of the 151 monthly rainwater collections on the whole period revealed a highly evaporated isotope line with respect to the GMWL, with a slope coefficient of 6.41 (r^2 = 91 %). Moreover, almost half (85 of 151 values) of the monthly data showed evaporated values with a deuterium-excess d < 10 ‰ regardless of the amount of monthly precipitation. The comparison between monthly and daily data over the same period revealed that monthly data present higher values and lower d than daily samplings. Thus, monthly data present a bias possibly due to the insufficient preservation of the rainwater during the monthly collection (evaporation and associated isotopic fractionation within the water collector during the monthly storage) and may not be related to evaporation processes during the water drop fall. Therefore, daily data appear to be more appropriate than monthly data to correctly define the isotope input signal.

The isotopic composition of rainwater has been therefore determined from the analysis of 212 daily rainfall samples, collected on 2 rainfall stations over the period from 23/03/ 2018–22/10/2019. The $\delta^{18}O$ values of precipitation ranged from –12.97 to +2.27 ‰, and the $\delta^{2}H$ values ranged from –98.5 to +20.2 ‰. This important variability of isotope ratios is a characteristic of tropical regions with strong seasonal contrasts [19]. Evaporated samples corresponding to minor rainfall events and due to undersaturation of the atmosphere were not taken into account to establish the local meteoric water line (LMWL; ≈13 % of the total data). The preliminary LMWL of Quixeramobim thus obtained from the daily monitoring network is defined by the following equation:

$$\delta^{2}H = 8.18*\delta^{18}O + 14.52 \quad (r^2 = 98\%, n = 184) \tag{1}$$

The slope coefficient of this partial LMWL is consistent with the global meteoric water line (GMWL). However, the y-intercept value of Equation (1) shows a d notably greater than +10 ‰. The proximity of the study zone to the Atlantic coast (≈ 190 km) and thus, to the oceanic moisture source, should imply a strong similarity between GMWL and LMWL. Nonetheless, IAEA-GNIP data obtained from the unevaporated precipitation chronic of Fortaleza (1965–1984 period, n=50) also showed a d value higher than the GMWL, equal to +12.7 ‰ (r^2 = 97 %). As a consequence, the high d value obtained from Equation (1) probably indicates that local precipitation partly results from a recycling of continental vapours induced by the evaporation of soils, plants or water bodies such as surface water [17]. A mixing of the oceanic moisture with recycled continental vapours which come from the numerous artificial or natural ponds is probably the first cause of observed high d value, but an occasional recycling with continental vapours from the Amazonian region could also be added to this first cause [17]. In any case, these two years of daily monitoring represent a first estimation of the input signal and measurements will be continued in order to validate the present definition of Quixeramobim's LMWL.

The annual mean isotopic composition of rainfall, weighted by rainfall amounts and determined from daily data obtained over the two years monitoring, is –3.21 ‰ for $\delta^{18}O$ and –12.0‰ for $\delta^{2}H$ (n=212). At the monthly scale, precipitation monitoring allows to observe a seasonal variability of isotopic compositions, with weighted monthly mean isotopic concentrations generally more depleted during the rainiest months (February to May; $r^2 \approx 0.5$; Table 1). The same behaviour was observed at Fortaleza during the period from 1965 to 1984 [20]. These monthly variations, in agreement with the classic pattern that controls the isotopic composition of precipitation during the year in the tropics, are related to the mass effect, which appears to be the main factor affecting the isotopic signature of precipitation in north-eastern Brazil [21]. In addition, the comparison of the isotopic composition of rainfall between the two stations enables the observation of an altitude effect, with more negative values for the Radar station, which is located 395 m of altitude above the Riacho Verde station (Table 1).

Table 1. Monthly weighted mean isotopic values of precipitation and monthly rainfall amounts obtained from the two monitoring stations (Riacho Verde and Radar).

	Date	01/18	02/18	03/18	04/18	05/18	06/18	07/18	08/18	09/18	10/18	11/18	12/18
RV	W. $\delta^{18}O$ mean (‰)	-	-	-3,09	-4,47	-2,54	-0,40	-0,36	-	-	-	2,27	-0,94
	Monthly Rainfall (mm)	-	-	70	249	88	25	8	0	0	0	3	64
RAD	W. $\delta^{18}O$ mean (‰)	-	-	-4,15	-4,49	-2,95	-1,16	-0,57			-0,53	-0,85	-1,30
	Monthly Rainfall (mm)	-	-	39	275	157	16	5	0	0	14	4	15
	Date	01/19	02/19	03/19	04/19	05/19	06/19	07/19	08/19	09/19	10/19	11/19	12/19
RV	W. $\delta^{18}O$ mean (‰)	-2,53	-3,09	-3,37	-3,54	-3,20	-0,92	-0,79	-	-	-	-	-
	Monthly Rainfall (mm)	132	248	150	105	100	56	67	0	0	-	-	-
RAD	W. $\delta^{18}O$ mean (‰)	-3,07	-3,58	-3,46	-5,08	-2,36	-1,74	-1,52	-	0.54	0.49	-	-
	Monthly Rainfall (mm)	172	134	95	191	78	20	99	0	1	2	-	-

4.2 Characterisation of groundwater recharge mechanisms

4.2.1. Hydrodynamics

Piezometric levels monitoring showed that high water levels are reached between May and July following the mean rainfall season, while the low water levels are reached at the end of the dry season around December/January (Figure 3). Generally speaking, our GW survey highlighted the high heterogeneity of the crystalline media, with EC ranging between 1059 and 14260 µS/cm, shallow water levels comprised between 0.76 and 21.7 m deep and seasonal variations of water level contrasted depending on boreholes (varying from 0.6 to 11.3 m).

The absence of DGPS levelling did not allow the development of piezometric maps, but the survey enabled to observe the high reactivity of the aquifer to precipitation and to the form of rainfall distribution (significant increase of the water table when the rainy events are significant or cumulative in a limited time; Figure 3). A number of boreholes showing strong seasonal variations (>2 m) are close to potential preferential infiltration zones (such as edges of surface water reservoirs, stream-channel beds, local depression zones, or even a recharge through the alluvium). The low soil thickness, the non-uniform grain size and the higher clay contents of the upper soil layers favour Hortonian-type flows, which leads to a predominant runoff compared to infiltration during intense rain or rain events concentrated over short periods of time. The geomorphology of the area and the presence of numerous ponds (natural or artificial) imply localised endorheism. Therefore, the recharge of the aquifer is probably favoured by the accumulation of water in these areas propitious to infiltration.

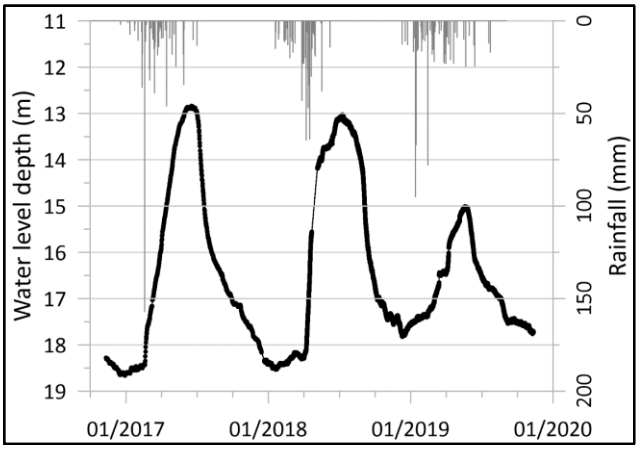

Figure 3. Seasonal fluctuation of water table depth (in black) in the crystalline basement aquifer. Daily rainfall (in grey) measured at Ibicuitinga Station (n°202).

4.2.2. Isotopic characterisation of groundwater

The isotopic characterisation of GW is based on the analysis of ^{18}O GW samples corresponding to 62 distinct boreholes sampled between 2009 and 2018, and on the analysis of 65 surface samples corresponding to the sampling of 10 dams between 2009 and 2012 and 3 dams in 2018. Analytical results of the GW stable isotopes are available in the Supplemental material, Table SM3. The isotopic composition of GW ranged from −4.19 to +1.76 ‰ for $\delta^{18}O$, −25.1 to +8.0 ‰ for $\delta^{2}H$, and −8.6 to +11.6 ‰ for d. Mean values and standard deviations are, respectively, of −2.20 ± 0.93 ‰ for $\delta^{18}O$, −13.3 ± 5.0 ‰ for $\delta^{2}H$, and 4.4 ± 3.3 ‰ for d. When plotting the isotope data of GW, surface water and daily precipitation on a $\delta^{18}O$ vs $\delta^{2}H$ diagram (Figure 4), we can observe that GW and surface water data are scattered along linear regression lines defined by the following equations:

- On one side, a linear regression line corresponding to GW data:

$$\delta^{2}H = 5.01*\delta^{18}O - 2.21 \ (r^2 = 90\%, n = 180) \qquad (2)$$

- On the other side, a linear regression line corresponding to surface water data:

$$\delta^{2}H = 4.92*\delta^{18}O - 2.31 \ (r^2 = 98\%, n = 65) \qquad (3)$$

Evaporation from surface water is obvious from its $\delta^{18}O$ values ranging between −1.32 and +9.23 ‰ and from the slope of 4.92 in Equation (3). Equation (3) thus symbolises the regional evaporation line. In the same way, GW samples are aligned along a line of slope 5.01 which indicates that meteoric waters have suffered evaporation prior to their infiltration into the aquifer [22]. Indeed, all GW data present isotope values marked by the presence of evaporated water if compared to the current definition of the LMWL (Figure 4). The strong similarity between Equations (2) and (3) highlights that GW recharge is mostly realised indirectly through a mixture process with evaporated surface water [8]. However, the intercept between the GW linear regression line and the LMWL of the $\delta^{18}O$ vs $\delta^{2}H$ diagram (Figure 4) shows an offset of about −2 ‰ in $\delta^{18}O$ compared to the present definition of the precipitation weighted annual mean (i.e. isotope input signal). This offset may correspond to the natural inter-annual variability of the weighted annual mean isotopic composition of rainfall, but could also highlight that only intense rainfall events from the rainy season (i.e. February to May) really participate to the GW recharge. This last hypothesis is supported by the GW level survey and by the weighted monthly mean isotopic composition most depleted during the rainiest months.

The spatial heterogeneity of GW isotopic composition existing between neighbouring boreholes (Supplemental material Figure SM1), which was already observed from GW survey, suggests that water wells explore isolated fracture systems and that the recharge processes occur locally. These observations support the fact that there is no regional subterranean flow within the Cearense crystalline aquifer [16]. Moreover, historical stable isotope data obtained from the 2009–2012 GW monitoring showed a seasonal variation of GW $\delta^{18}O$ of about −1.03 to −3.90 ‰ depending on the borehole. Therefore, seasonal or (inter)annual variability of GW isotope signal of most boreholes suggests rapid recharge and circulation processes within the aquifer.

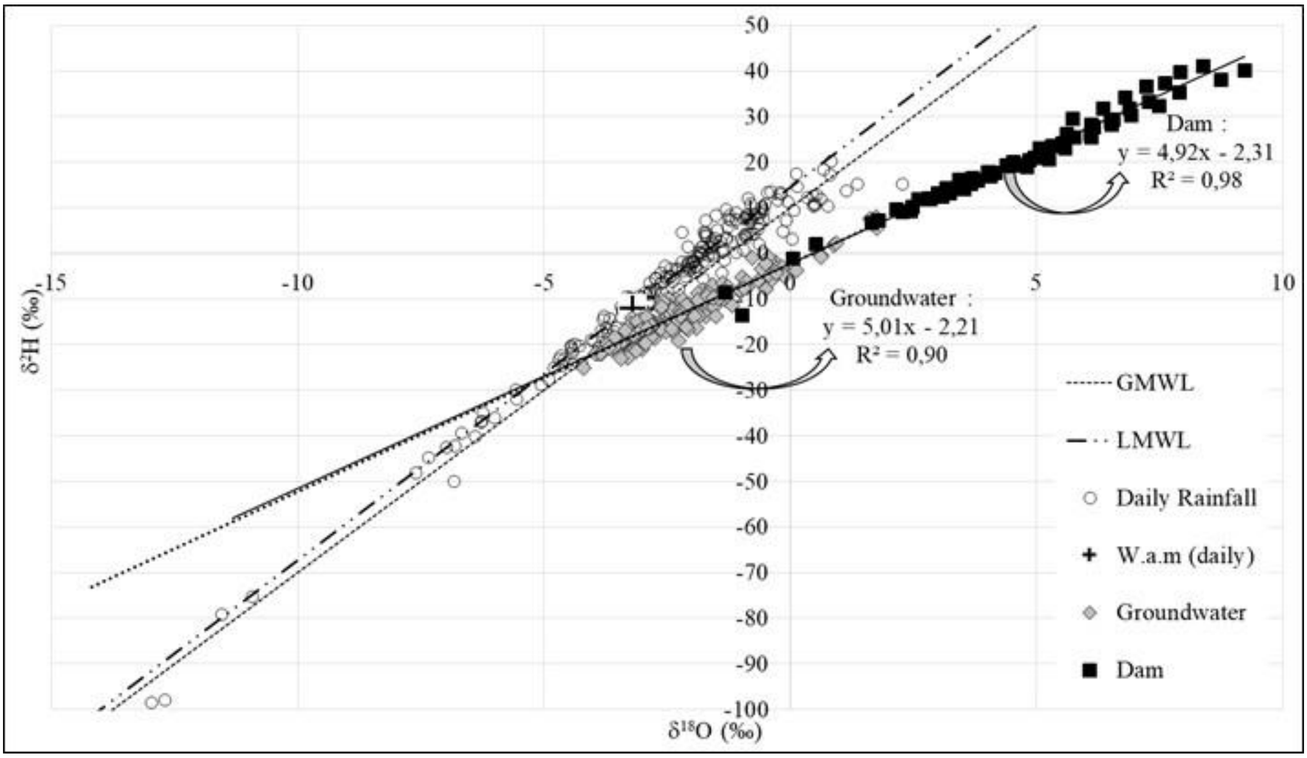

Figure 4. Isotopic composition of groundwater (grey diamonds), surface water (black squares) and daily rainfall (white circles).

4.2.3. Groundwater residence time

Estimation of GW residence time via a multi-tracer approach (^{14}C, ^{3}H, CFC, SF_6) allowed to evaluate the apparent age of GW. Indeed, ^{14}C and ^{3}H analyses highlighted a large contribution of post-1950s water with values of ^{14}C generally higher than 100 pMC and a measurable presence of tritium (≤0.3 < ^{3}H < 1TU; Table 2).

However, CFC and SF_6 analyses showed that the most coherent model for almost all samples was a binary mixture model (BMM; of the piston flow type) between an old end-member (<1960) and a present-day end-member (Supplemental material Table SM4), consistent with ^{14}C and ^{3}H data. The BMM suggests different GW flow path organisation with variable contributions between a fast vertical flow (present-day end-member; 15–85 %) and a slower horizontal flow (old end-member). The strong contribution of present-day recharge validates the hypothesis of rapid recharge and circulation processes. A basic conceptual model of the processes is proposed in Figure 5.

Table 2. Analytical results of 14C and 3H in groundwater.

	Point	Date	^{14}C (pMC)	^{3}H (TU)
Forquilha	FOR 144	12/2017	103.6 ± 0.5	0.6 ± 0.3
	FOR 151	12/2017	98.3 ± 0.5	≤ 0.7
	FOR P7	11/2009	126.7 ± 0.5	0.7 ± 0.6
Ibicuitinga	IBI 160	12/2017	91.8 ± 0.5	≤ 0.3
	IBI 96	12/2017	73.2 ± 0.6	≤ 0.4
Pirabibú	PIR 52	12/2017	105.6 ± 0.5	≤ 0.5
	PIR P16	12/2017	105.3 ± 0.5	0.7 ± 0.3
	PIR 49	11/2009	124.4 ± 0.6	1 ± 0.4
Vista Alegre	VA 132	12/2017	110.1 ± 0.5	≤ 0.7
	VA PPz	11/2009	119.5 ± 0.5	0.9 ± 0.5

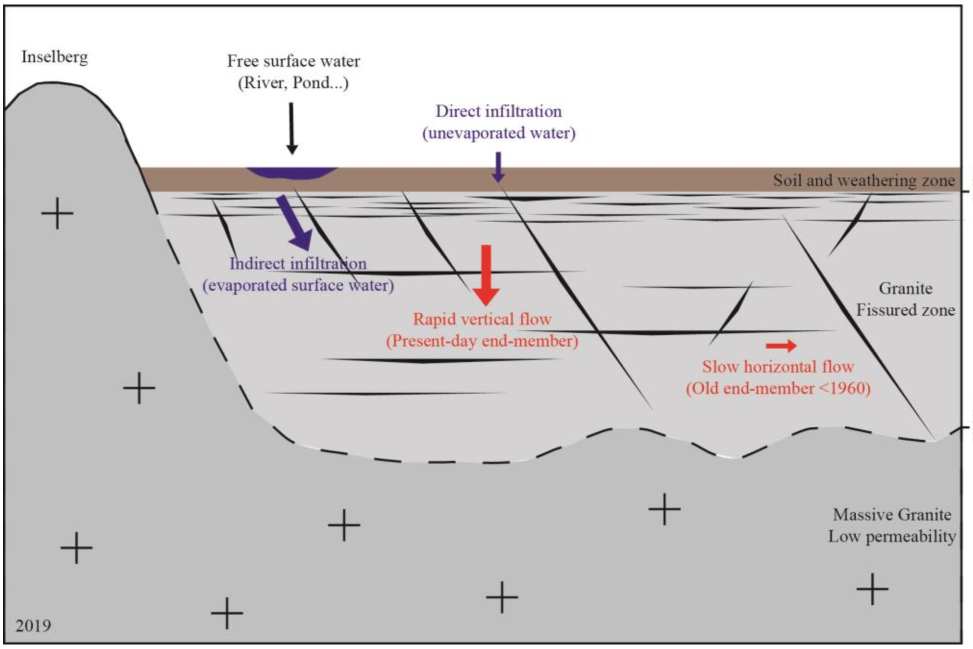

Figure 5. Basic conceptual model of the recharge processes and time residence of the groundwater contained in the fractured crystalline aquifers of semi-arid Ceará. The time residence data is based on the year 2019. Large arrows correspond to a dominancy of the process regarding to the other.

5. Conclusions

This study provided new insights into the hydrogeosystems functioning and the recharge mechanisms of the shallow fractured crystalline aquifers of Ceará. Indeed, hydrodynamic data highlighted the high reactivity of aquifers to precipitation and to the form of rainfall distribution, with a strong regular increase of the water table when the rainy events are significant or cumulative in a limited time. The fast raising of GW levels, combined with geomorphological and pedological information, suggests that the infiltration of waters occurs principally through preferential infiltration zones. The analysis of stable H and O isotopes allowed to specify that Precambrian basement aquifers of the semi-arid region of Ceará are mainly recharged by meteoric waters through indirect infiltration from evaporated surface water. The GW residence time determination, through a multitracer approach, has provided useful information about the spatio-temporal heterogeneity of flow path organisation within aquifers, with variable contributions between a fast vertical flow (present-day end-member; 15–85%) and a slower horizontal flow (old end-member <1960). In consequence, hydrodynamic and hydrogeochemical data suggest that crystalline basement aquifers are rapidly recharged by evaporated surface waters through preferential infiltration zones (i.e. low points of the relief). The seasonal variability of the GW isotopic composition presupposes that the mixing proportion between direct and indirect infiltration varies over the year, once again emphasising the spatio-temporal heterogeneity of these processes. This research proves that local aquifer systems are highly heterogeneous, but well connected to the land surface. This implies that aquifers are very sensitive to drought and pollution, which are important parameters for the sustainable management of these water resources.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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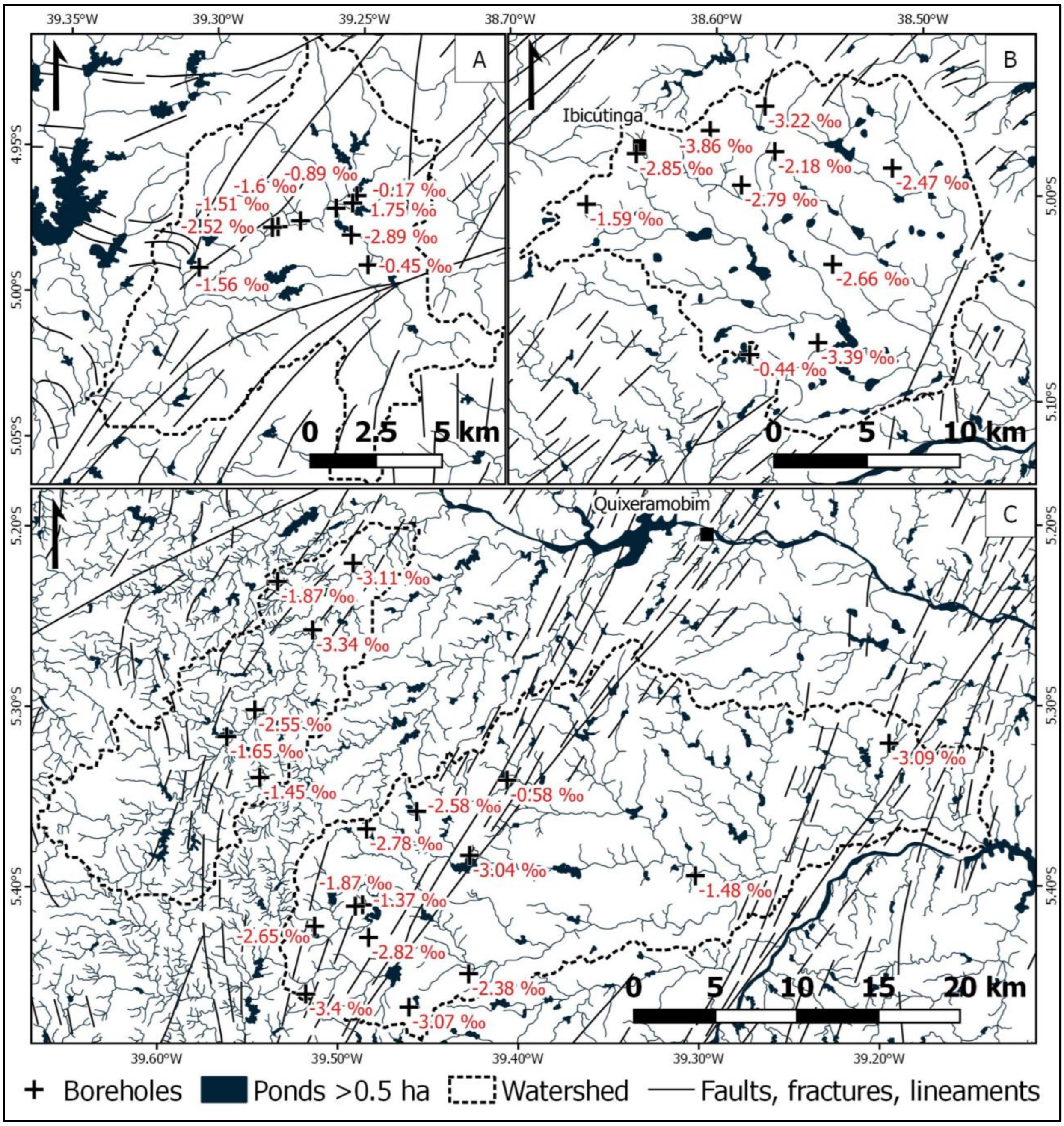
Supplementary material



Supplemental material Figure SM1. Groundwater δ¹⁸O composition (June 2018 campaign). The boreholes labels correspond to the δ¹⁸O value obtained from the campaign of June, 2018. A) Map of Pirabibú watershed. B) Map of Ibicuitinga watershed. C) Map of Forquilha (left side) and Vista Alegre (right side) watersheds.

Supplementary material Table SM1. Characteristics of studied watersheds.

	Forquilha	Pirabibú	Vista Alegre	Ibicuitinga
Area	214 km²	127 km²	550 km²	286 km²
Municipality	Quixeramobim			Ibicuitinga and Morada nova
Mean annual precipitation [1981-2010]	688 mm			749 mm
Maximum altitude	682 m	761 m	545 m	267 m
Minimum altitude	205 m	216 m	132 m	39 m
Average slope	9.2%	-	-	-
Geomorphology	Undulating relief	Undulating relief in the upper part - slightly undulating relief in the lower part	Slightly undulating relief and presence of residual massifs	Flat area and low incision of the local drainage network in the landscape.

Supplementary online material Table SM2. Description of the five rainfall stations.

	Monthly monitoring			Daily monitoring
Station Name	Inmet-Quixeramobim	Algodões-ZéNobre	Vista Alegre	Radar
Station ID	-	-	-	766
Station type	Experimental			Conventional
Municipality	Quixeramobim			
Latitude (WGS84)	-5.17	-5.38	-5.34	-5.07
Longitude (WGS84)	-39.29	-39.58	-39.40	-39.27
Altitude	208 m	289 m	206 m	663 m
Distance to the coast	172 km	212 km	195 km	165 km

Supplementary material Table SM3. Distribution of groundwater δ^{18}O composition.

	Forquilha	Ibicuitinga	Pirabibú	Vista Alegre	Banabuiú
N° of samples	31	22	47	39	41
N° of boreholes	8	11	12	17	14
δ^{18}O min. (‰)	-4.19	-3.86	-3.43	-3.57	-3.68
δ^{18}O max. (‰)	-0.86	-0.44	1.75	1.76	-0.49
δ^{18}O median (‰)	-3.08	-2.94	-1.60	-2.54	-2.81
δ^{2}H min. (‰)	-25.06	-22.02	-23.03	-20.56	-21.44
δ^{2}H max. (‰)	-7.46	-5.47	7.96	5.56	-7.26
δ^{2}H median (‰)	-18.28	-14.82	-10.46	-13.75	-16.36
d min. (‰)	-2.02	-1.98	-6.06	-8.55	-3.98
d max. (‰)	9.78	11.57	7.42	10.12	10.13
d median (‰)	6.35	7.86	2.25	6.18	4.46

Supplementary material Table SM4. Analytical results of the CFC and SF_6 dating tracers and deduced end-members of the Binary Mixture Model (piston flow type). “Corrected SF_6” correspond to the SF6 value corrected by the excess air amount of the sample. “*” Samples presenting a SF_6 contamination.

Point	Corrected SF6 (pptv)	CFC12 (pptv)	CFC-11 (pptv)	CFC-113 (pptv)	Binary Mixing Model (piston flow type)			
					Recent pole (%)	Recent age (year)	Old pole (%)	Old age (year)
FOR 144	6.2	330.2	111.2	42.4	65	0	35	>60
FOR 151	4.1	368.9	141.9	58.4	50	0	50	30-40
IBI 89	2.0	-	98.8	35.5	40	15	60	>60
IBI 96	34.5*	11.0	35.4	13.7	20	0	80	>60
IBI 152	19.6*	68.5	41.4	13.7	15	0	85	>60
PIR 44bis	11.1	550.9	172.6	78.9	-	-	-	-
PIR 45	8.3	400.3	133.7	55.5	85	0	15	>60
PIR 52	4.3	133.4	67.8	26.4	35	0	65	>60
VA 123	3.0	174.5	104.6	46.0	25	0	75	>40
VA 132	4.2	155.6	106.9	29.6	40	0	60	>60