



Assessment of geochemical processes in the shared groundwater resources of the Taoudeni aquifer system (Sahel region, Africa)

Rim Trabelsi¹ · Kamel Zouari¹ · Luis J. Araguás Araguás² · Adnane S. Moulla³ · Aboubacar M. Sidibe⁴ · Teis Bacar⁵

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Abstract

The Taoudeni basin occupies a large portion of the West African Craton in Mauritania, Mali, SW Burkina Faso, and Algeria. The multi-layer Taoudeni aquifer system (TAS) occurs in geological formations extending in age from the Infra-Cambrian to the Quaternary. These groundwater resources support local livelihoods, maintain vital ecosystems, and strongly affect terrestrial water and energy budgets in the region. Hydrogeochemical and stable ($\delta^2\text{H}$, $\delta^{13}\text{C}$, $\delta^{18}\text{O}$) and radioactive (^3H , ^{14}C) isotope analyses were carried out with the aim of identifying the major geochemical processes controlling groundwater quality and the recharge mechanisms. The analyzed groundwater samples showed a wide range of chemical compositions based on major ions, with limited variability, regardless of the aquifer lithology or stratigraphy. The predominant water type, Mg–Ca– HCO_3 , was found in most geological and hydrogeological settings. A principal component analysis of the hydrochemical data revealed the two major processes governing the geochemistry of groundwaters in the TAS. These are water–rock interaction (leaching, weathering, ion exchange) and anthropogenic contamination. Environmental isotopes revealed the presence of groundwaters with heterogeneous signatures, reflecting different recharge processes and varying groundwater ages. A widespread present-day recharge by direct infiltration of rainwater and/or by surface water was highlighted by significant tritium levels in most wells and high ^{14}C activities occurring mainly in shallow aquifers. However, isotopically depleted $\delta^{18}\text{O}$ contents with low ^{14}C activities were found in the confined part of the Continental Intercalaire aquifer, particularly in the Algerian part, indicating the presence of fossil groundwater, recharged under colder past climatic conditions.

Keywords Arid regions · Isotopes · Water–rock interaction · Groundwater contamination · Africa

Introduction

In the last few years, the strategic importance of groundwater resources in Africa has been increasingly recognized, particularly their importance for community water supply. The sustainable use and protection of these valuable resources are considered crucial to achieving the SDG6 Development Goal aiming to ensure access to clean water, sanitation and hygiene, which are the most basic human needs for health and well-being. Nevertheless, the management of water resources in Africa, and in particular in the Sahel region, faces significant challenges from quantitative and qualitative viewpoints. The increase in water demand linked to population growth and economic development comes up against the decreased availability of groundwater due to the impacts of climate change and the often-inefficient management of water resources. Furthermore, water quality degradation has reached worrying proportions due to highly polluting practices.

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✉ Rim Trabelsi
trabelsi.rim@enis.tn

¹ Radio-Analysis and Environment laboratory, ENIS, Sfax University, Sfax, Tunisia

² Isotope Hydrology Section, International Atomic Energy Agency, Vienna, Austria

³ Algiers Nuclear Research Centre, PO Box 399 Alger-RP, 16000 Algiers, Algeria

⁴ Ministry of Energy and Water, Direction Nationale de l’Hydraulique, Bamako, Mali

⁵ Ministry of Hydraulics and Sanitation, Centre National des Ressources en Eau, Nouakchott, Mauritania

Improving overall water management in arid and semi-arid regions is now a key priority to help achieve the goals of poverty alleviation and sustainable development. In particular, insufficient knowledge and poor management of groundwater sources in urban and rural areas need to be addressed. This is the case for almost all of the West Africa water resources that are shared between several nation states, both at the level of the hydrological basins (e.g., Niger, Senegal, Volta, Gambia, etc.) and aquifer systems. The Taoudeni basin contains one of the most important aquifer systems in the Sahel region. It is shared between Mali, Mauritania, Algeria, and Burkina Faso. This basin presents a significant volume of groundwater resources (about 11 billion m³).

However, the current climatic conditions in the Taoudeni basin, varying from a hot and humid climate in the southern part to an arid climate in the central and the northern sectors, do not allow complete replenishment of the water resources extracted to cover the growing demand. In the northern part of the Taoudeni basin, the population density is low, and water consumption is thus low. However, in the central part, especially in the southern area, the population growth rate is about 3%/year. The population has continued to grow at a very high rate, with an increase of about 10 million people since 1990 (World Bank 2020). Furthermore, higher temperatures are expected to occur in this region due to climate change, which will exacerbate extreme climate events of droughts and floods (AU 2020; Nangombe et al. 2018). These processes could reduce the natural recharge processes of this aquifer, increase evaporation losses and affect water quality.

In this context of high population growth, food insecurity and the impacts of climate change, water resources face several critical challenges in the Taoudeni basin, namely ensuring access to good quality water for the entire rural and urban population, despite the projected disruption to rainfall patterns.

Thus, groundwater resources play a vital role in the Taoudeni basin, greatly contributing to the socio-economic development of the region. This situation involves increasing groundwater abstraction, resulting in pronounced lowering of hydraulic heads in several aquifers, and water quality degradation as a result of human activities (e.g., lack of sanitation), and also poses risks with respect to environmental protection. Unfortunately, the hydrological and geochemical processes that govern groundwater recharge, sustainability, and sensitivity to climatic variability are poorly understood. Determining the geochemical processes that control groundwater quality is vital for better assessment and effective management of the groundwater resources (Toth 1999; Edmunds et al. 2003; Chen et al. 2004). Groundwater quality is mainly affected by the geological formations, changes in land use, and the geomorphology of the studied regions (Kelepertsis 2000; Siegel 2002; Prasanna et al. 2010; Khmila et al. 2021).

Variability in the aquifer matrix plays an important role in the hydrochemical evolution of groundwater quality depending on the reactivity of the predominant rock types and on the timescales of flow and geo-microbiological interactions.

In such context, chemical tracers are commonly used to investigate these geochemical processes, since groundwater chemistry reflects the lithology in the recharge areas and hence helps to unravel the flow path evolution (Edmunds et al. 2003; Chidambaram et al. 2011; Thilagavathi et al. 2012; Thivya et al. 2013, Trabelsi and Zouari 2019).

Additionally, environmental isotopes have been widely applied as natural tracers in hydrological and hydrogeological investigations. These tracers are particularly effective for conducting groundwater investigations in semi-arid and arid regions. They are helpful for identifying the groundwater origin and main flow paths, assessing mixing patterns between different groundwater bodies, and estimating the residence time of groundwater in the aquifers (Fontes 1980; Clark and Fritz 1997; Cook and Herczeg 2000; Etcheverry 2002; Trabelsi et al. 2020).

Numerous studies and projects in hydrogeology and water quality (SOGREAH 1988, 1994; IWACO 1989; Ouedrago 1994; Dakouré 2003, 2010; Huneau et al. 2011) have focused on the eastern part of the Taoudeni basin, intending to characterize local groundwater in terms of hydrodynamic, geochemistry and residence times. This investigation was mainly based on the isotopic and chemical contents of groundwaters sampled in various units of the Taoudeni aquifer system (TAS) within the framework of two regional technical cooperation projects supported by the International Atomic Energy Agency (IAEA). The combination of data on the major chemical elements and several stable and radioactive isotopes was used in conjunction with principal component analysis (PCA) methods to characterize groundwater quality and to define the dominant geochemical processes, as well as to identify and characterize the recharge processes.

Regional settings

The Taoudeni Basin is a major geological sedimentary basin in West Africa, extending over 2,000,000 km². It covers large parts of the African Craton in Mali (57%), Mauritania (25%), Algeria (16%) and the southwestern part of Burkina Faso (2%) (OSS 2007; Derouane 2008; Fig. 1).

This basin is characterized by irregular precipitation patterns, varying from less than 100 mm/year in the northern sectors to more than 1,200 mm/year in the southern sector. Thus, four climatic zones are identified:

- The Sudanian zone with a Guinean climate and annual rainfall greater than 1,200 mm, and sometimes 1,500 mm

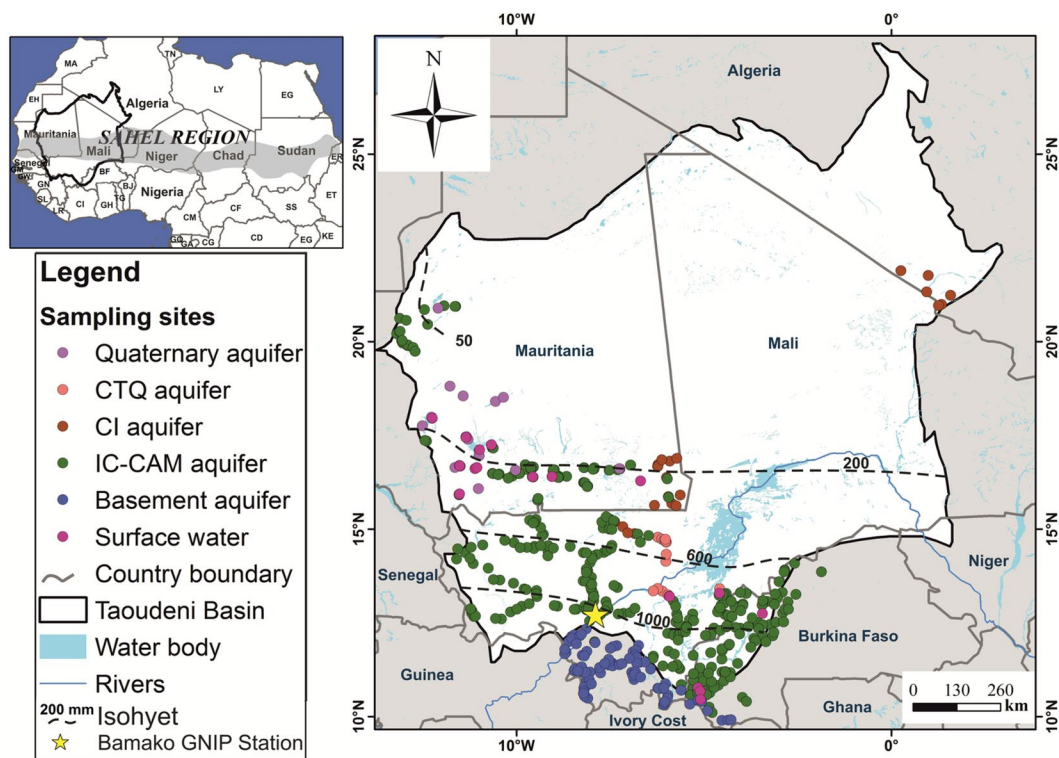


Fig. 1 Location map of the Taoudeni basin, showing the positions of the water sampling sites and the spatial distribution of annual rainfall

- The Sudano-Sahelian zone with a tropical climate and annual rainfall ranging from 600 to 1,200 mm
- The Sahelian zone with annual rainfall ranging from 200 to 600 mm
- The sub-Saharan zone with a subdesert type climate and annual rainfall varying from less than 50 to 200 mm

The spatial distribution of the precipitation in the Taoudeni basin is the inverse of the potential evapotranspiration (ETP) distribution. The mean annual relative humidity does not exceed 50% in the north and increases gradually in the southern part of the TAS. The minimum ETP is typically recorded from February to March and the maximum from June to October. The mean annual air temperatures are high, ranging from 35 to 45°, and the maximum could reach 51 °C in the northern sector of the basin. Temperatures generally increase from the southwest to the northeast (Gerbe 2013). Analysis of the annual temperature and rainfall variations with time reflects the existence of global warming since 1960, with an average rate of 0.15 °C per decade and a general decrease in the precipitation amount. Although the decline in rainfall can be expressed as a steady decline, there is considerable year-to-year variability and evidence of cyclical trends. Extreme droughts have occurred in the recent past (the 1940s and from 1980 to 1995), with relatively wetter

periods including the 1950s and the 1990s onward (World Bank 2011; Niang et al. 2014; USAID 2018). This intra-annual variation suggests a significant temporal variability of the aquifer's recharge.

Geology and hydrogeology

The Taoudeni is the most prominent sedimentary syncline basin in the West African craton, which was formed during the mid-late Proterozoic. It is bounded to the north by the Reguibat Shield, to the south by the West African Shield craton and to the east by Iforas and Hoggar crystalline massifs. The stratigraphic succession of the Taoudeni basin extends from the Proterozoic/Palaeozoic units to the Quaternary sediments. The basin represents one of the world's most widespread Proterozoic successions and contains up to 6,000 m of Late Precambrian and Palaeozoic sediments (Rooney et al. 2010). The Mesozoic–Cenozoic units form a thin cover in the basin centre, which outcrops under lacustrine systems and Quaternary sand and dunes.

The structural evolution of the basin suggests epeirogenic movements, which generated low angular unconformities, and the reactivation of basement faults frequently injected by diabase dikes (Shields et al. 2007). These normal faults

created a structure of horsts and grabens across the basin in the Precambrian basement and associated syn-sedimentary breccias in the lower part of the sedimentary unit (Fig. 2). The deposits of the Mesozoic–Cenozoic to the Quaternary units are generally devoid of tectonic features (Shields et al. 2007). These last deposits show a spatial continuity between different basins, Taoudeni and Tanezrouft basin in the northern part and Taoudeni and the Iullemeden basin in the eastern sector.

Controlled by the geologic structure, the extensive aquifer system in the Taoudeni basin includes different geological formations ranging in age from the Precambrian to the

Quaternary. The Taoudeni aquifer system is often subdivided into very distinct superimposed sedimentary units. The first unit is composed of a set of ancient sedimentary terrains of Paleozoic age, surmounting the Precambrian folded basement made up of consolidated rocks and discontinuous fissured aquifers. The second unit is represented by recent sandstone-dominated sedimentary terrains ranging from the Mesozoic to the Quaternary, consisting of several continuous aquifers that extend over the entire basin. The identified aquifer formations in the Taoudeni basin are (Fig. 2; Table 1):

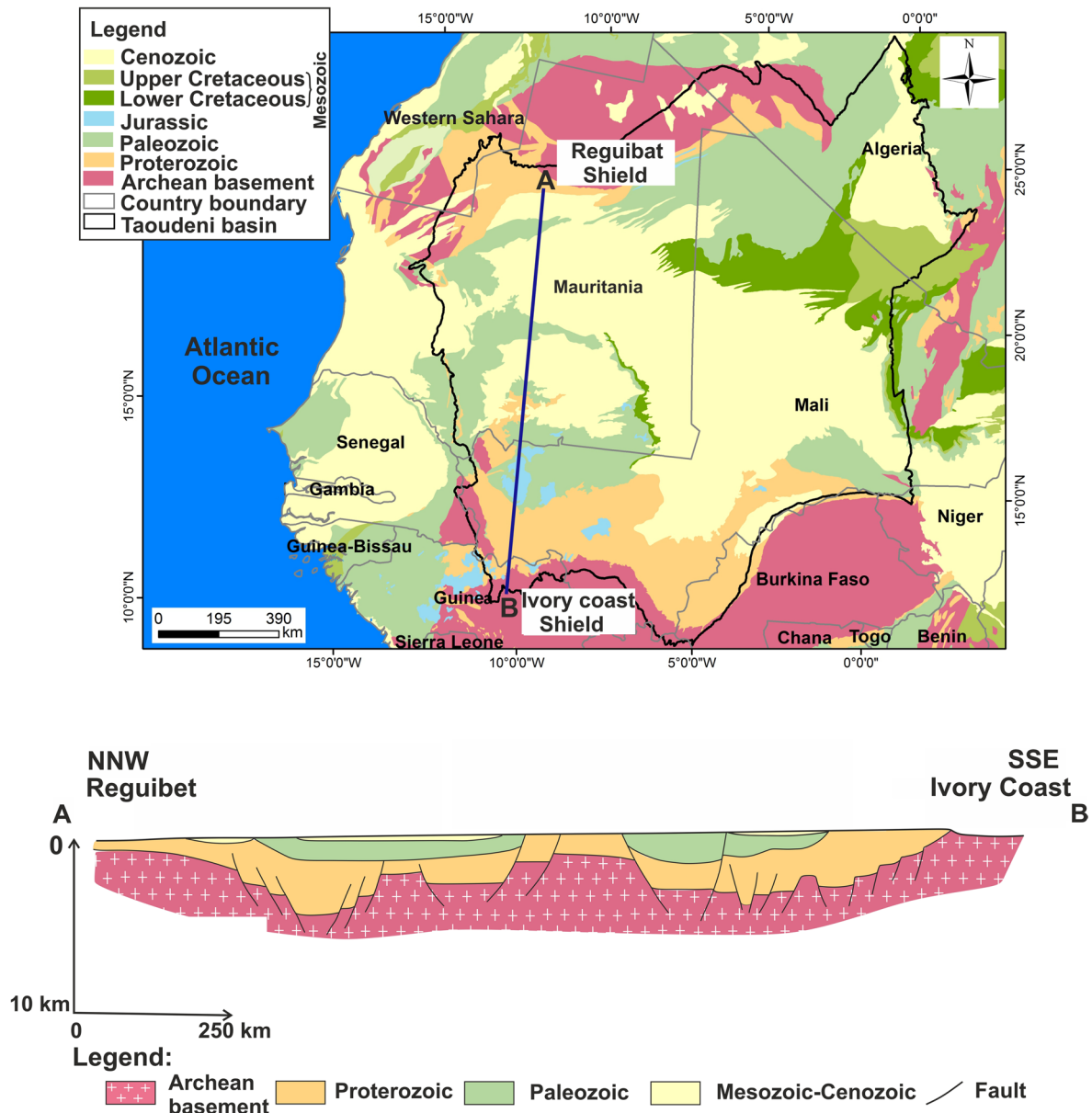


Fig. 2 Geological map of the Taoudeni basin and north–south simplified cross-section showing the stratigraphic structure of the basin (after Albert-Villanueva et al. 2016)

- **Basement:** Composed of granites, mica schists, and shales of the Precambrian and is exploited mainly in Mali, where it is affected by several families of fractures.
- **Infra Cambrian (IC):** Constituted by consolidated sedimentary rocks outcropping on a large area in the south of Mali and the south-west of Burkina Faso (BKF) and forms discontinuous aquifers. In Mauritania, the IC is composed of the Ayoune sandstones. In Mali, the most exploited layers are the tabular (ICT) formations of the Infra Cambrian and the folded ones (ICP). In Burkina Faso, the IC units infill unconformably into the crystalline basement. It is essentially made of sandstone, with locally more or less carbonated cement composed of nine units.
- **Cambrian (CAM):** Comprises thick sand and very fine-grained silica-clay rocks (pelites) in Mali and Mauritania. The groundwater potential of this aquifer depends on the predominance of schistose formations in the Cambrian series and the frequency of the doleritic intrusions, which are often massive.
- **Continental Intercalaire (CI):** Formed by sands and sandstones of the Upper Cretaceous and lower Eocene ages and located in Algeria, Mauritania and Mali.
- **Continental Terminal (CT):** Composed of lacustrine fluvial deposits of the upper Eocene to Oligocene, containing alternating sand and clay layers, with a thickness of about 40 m. In some sectors of the basin, the lithological facies of the CI and CT formations are similar and difficult to differentiate. The CT overlays in discordance the lower parts of IC formations. The groundwater flow direction is mainly along the SW–NE direction.
- **Quaternary:** These sediments constitute the shallowest aquifer that is discontinuous and composed of recent sandy-clayey alluvium along the main watercourses. The Quaternary aquifer could be associated with the

CT to form the Continental Terminal and Quaternary (CTQ) aquifers in Mali and Mauritania.

The most stressed aquifers in the Taoudeni basin are those of the Quaternary alluvium and the Continental Terminal, particularly in Mali (extraction greater than 30 Mm³/year). This observation is explained by the shallow nature of the aquifers, which facilitates access to groundwater. Deep boreholes tap aquifers in artesian conditions. Given their high cost, these deep boreholes are only justified for the mining industry; moreover, the exploitation of the Continental Intercalaire and the Continental Terminal is intense in Mali and very intense in Mauritania. The exploited groundwater resources are used for drinking water supply in urban areas, for livestock, as well as for irrigation and energy. The northern part of the Taoudeni basin (eastern region of Mauritania, northern Mali, and southern Algeria called Tanezrouft basin) is sparsely populated; the water points are generally located along the principal axes of transhumance.

Unfortunately, the flow directions of these different aquifers have not been studied in detail in previous hydrogeological studies which indicate that the regional groundwater flow is from the south east, the north west and the north east towards hydraulic head depressions that exist in the central part of the basin (Derouane 2008; Huneau et al. 2011; OSS 2013; Collignon 2022).

Due to the geographical extent of the study area and the complex hydrogeological setting, the selected aquifers to be studied in this paper are simplified as follows: (1) the Continental Intercalaire (CI); (2) the undifferentiated Continental Terminal and Quaternary aquifers (CTQ); (3) the Cambrian and Infra Cambria aquifers, which are joined together under the name of IC-CAM; (4) the Birrimian basement; and (5) the shallow aquifer made up of alluvial deposits of the Quaternary (Q) (Fig. 2).

Table 1 Litho-stratigraphic table of different geologic units of the Taoudeni basin, indicating the aquifers/aquitards

Unit age	Description	Aquifer/aquitard
Quaternary	Sandy-clayey alluvium/Sand dune	Q aquifer
Lower Eocene–Pliocene (Tertiary)	Unconsolidated sandstones and sands/Lacustrine fluvial deposits	CT aquifer
Upper Cretaceous–lower Eocene (Tertiary)	Marine sediments	Aquitard
Lower Cretaceous	Continental deposits of sand and sandstones	CI aquifer
Permian–Triassic	Dolerites, volcanic rocks composed of basalts and gabbros	Aquitard
Cambrian–Carboniferous	Sandstone, limestone and shale	CAM aquifer
Infra-Cambrian	Metamorphosed sandstone	IC aquifer
Precambrian	Granites, mica schists, and shales	Basement aquifer

Materials and methods

Sampling and analysis

All the chemical and isotopic analyses reported here were carried out within the framework of two regional technical cooperation projects supported by the International Atomic

Energy Agency (IAEA). Most of the sampling points are used as irrigation and drinking water supply wells of rural villages, and the depths vary from 2 to 150 m (Table 2).

A total of 429 water samples were collected to study the chemical properties of surface and groundwaters in the Taoudeni basin, including the Quaternary aquifer (20 samples), CTQ aquifer (16), CI aquifer (26), IC-CAM aquifer

Table 2 Range, mean values and standard deviations (SD) of the physical-chemical parameters and environmental isotopes measured in the different aquifer units of the Taoudeni Aquifer

Parameter	Aquifer CI				Aquifer CTQ				Aquifer IC-CAM			
	Mean	Min	Max	SD	Mean	Min	Max	SD	Mean	Min	Max	SD
Depth (m)	-	19	112	-	-	2	70	-	-	9	150	-
Temp (°C)	31.8	25.4	43.6	3.4	31.1	29.3	36.6	2.2	30.3	21.9	37	2.2
EC (µS/cm)	1,064.4	25.6	3,927	1,242.3	212.1	30.7	953	262.4	509.1	2.5	3,275	571.7
pH	7.2	6.2	8.5	0.7	7.5	6.5	8.1	0.4	7.1	3.6	9.9	1
δ ¹⁸ O (‰ VSMOW)	-6.5	-9	-1.9	2	-2.5	-6.3	1.3	2.5	-4.8	-7.8	1.6	1.1
δ ² H (‰ VSMOW)	-48	-67.4	-18.6	13.3	-20.4	-40.6	1	14.1	-32.9	-53.3	-1.7	6.5
δ ¹³ C (‰ VPDB)	-7.7	-12.7	-3	2.9	-10.3	-10.3	-10.3	-	-10.4	-16.5	-5.9	2.2
¹⁴ C (pmc)	42.4	1.7	86.1	34.2	20.2	20.2	20.2	-	81.2	15.6	116.8	22.4
³ H (TU)	0.9	0	2.9	0.8	2.6	0.5	6.3	1.7	2.1	0	9.1	1.8
TDS (mg/L)	542.2	17	2,945	726.1	191.4	23	681.1	180	333.4	8	2,810	436.4
Ca (mg/L)	61	1.6	450.4	98.8	16.5	1	58.8	16.9	33.6	0.5	445.7	50.5
Mg (mg/L)	19.6	0	92.1	26.1	9.4	0.3	35.6	10.8	15.8	0	142.6	20.8
Na (mg/L)	102.2	0.6	530	156.9	14.2	2.6	46.4	12.7	34.7	0.6	550.9	66.9
K (mg/L)	15.2	1.1	158.8	31.2	11.3	0.9	130.8	31.9	7.9	0	189.9	18.1
Cl (mg/L)	122.3	0.9	1,180.2	280.5	6.2	0.9	44.1	10.5	26.6	0	495	60.5
SO ₄ (mg/L)	124.9	0.3	616.8	176	14.7	0	186.5	46.3	37.8	0	1,341.3	120
NO ₃ (mg/L)	40.8	0	278.5	69.8	11.1	0	81.3	20.8	59.4	0	1,127.8	136.2
HCO ₃ (mg/L)	109.7	11	347.7	107.4	104.6	6.1	268.4	77.6	115.2	2.1	884.5	109.4
Parameter	Aquifer Basement				Aquifer Quaternary				Surface water			
	Mean	Min	Max	SD	Mean	Min	Max	SD	Mean	Min	Max	SD
Depth (m)	-	20	79	-	-	9	150	-	-	-	-	-
Temp (°C)	29.3	25.1	32.2	1.8	27	17.9	39.4	4.1	21.7	14.8	28.4	3.5
EC (µS/cm)	259.3	12.8	1364	254.6	865.6	178	3,331	736.3	195.4	28	755	181.3
pH	7.3	5	8	0.5	7.7	5.9	8.9	0.6	8	7	8.8	0.7
δ ¹⁸ O (‰ VSMOW)	-5.4	-6.5	-3.5	0.6	-4	-6.5	-1.2	1.5	10.3	-6.6	29.7	12.3
δ ² H (‰ VSMOW)	-35	-44.8	-20.1	4.5	-30.3	-44	-16.5	8.4	38.3	-41.9	130.2	56.1
δ ¹³ C (‰ VPDB)	-13.3	-17.7	-8	2.8	-10.6	-13.1	-9	1.8	-	-	-	-
¹⁴ C (pcm)	80.4	51.8	97.6	13.4	95.7	59.8	110	24.1	-	-	-	-
³ H (TU)	0.8	0	3.8	0.8	2.7	0	5	1.4	4.1	1.7	6.7	1.5
TDS (mg/L)	195.3	10.2	925.1	173.3	527.6	120.1	1,370.7	354.2	167.1	25.2	644	150.5
Ca (mg/L)	17.8	0.6	140.4	23.6	60.3	13.9	180.6	52.7	20.8	2.2	80.2	18.9
Mg (mg/L)	10.2	0	58.8	11	21.9	3.8	54.8	13.4	8.8	1.1	38.9	9
Na (mg/L)	16.3	1.3	91.9	13.4	65.7	9.4	167.1	57.1	12.2	1.4	69.1	18.7
K (mg/L)	4.5	0	31.3	4.8	16.9	2.9	35.5	10.1	11.7	1.3	28.5	9.5
Cl (mg/L)	7.6	0	169.4	25.5	61.5	4.8	302	78.9	9.1	0.6	45	11.6
SO ₄ (mg/L)	2.4	0	38.7	7.1	74.9	9	482.6	111.9	15.8	0	169.5	41.1
NO ₃ (mg/L)	19.1	0	335.0	64.1	47.6	0	268.4	61.3	5.5	0	15.7	5.1
HCO ₃ (mg/L)	117.4	4.1	274.5	63.2	222.4	36.6	329.4	66.3	107.5	18.3	280.6	79.5

(264), basement aquifer (86), and surface waters (17). The samples were collected, after purging the wells, in 1000 ml polyethylene bottles which were rinsed first with the sample's water several times and then filled to the top. Physicochemical parameters, including temperature, pH, and electrical conductivity (EC), were measured in the field. Major chemical elements (Na^+ , Ca^{2+} , Mg^{2+} , K^+ , Cl^- , NO_3^- , SO_4^{2-}) concentrations were determined using high-performance liquid ion chromatography (HPLC). The total alkalinity (as HCO_3^-) was determined at the laboratory by titration using 0.1 N HCl acid. The ionic balance for all samples was within $\pm 5\%$.

For water stable isotope analyses, 549 water samples were collected from surface and groundwater bodies in the Taoudeni basin as follows: Quaternary aquifer (24), CTQ aquifer (16), CI aquifer (24), IC-CAM aquifer (375), basement aquifer (91) and surface waters (17). Oxygen-18 and deuterium contents were measured either by laser absorption spectrometry (LGR DLT 100) equipped with an autosampler or by the CO_2 equilibration method for $\delta^{18}\text{O}$ (Epstein and Mayeda 1953) and the isotopic exchange method with H_2 gas for $\delta^2\text{H}$ (Coplen et al. 1991), followed by mass spectrometry measurements. The isotopic results are reported using δ notation relative to the Vienna Standard Mean Ocean Water (‰ versus VSMOW standard). The analytical uncertainty of the reported values is about $\pm 0.2\text{‰}$ for $\delta^{18}\text{O}$ and $\pm 1\text{‰}$ for $\delta^2\text{H}$ (one-sigma level).

A total of 340 water points from surface water and groundwater were selected for tritium analysis in the Taoudeni basin: Quaternary aquifer (21), CTQ aquifer (15), CI aquifer (17), IC-CAM aquifer (221), basement aquifer (49), and surface waters (17). Tritium analyses were performed by liquid scintillation counting (Taylor 1976) after electrolytic enrichment. Tritium concentrations are expressed in tritium units (TU), which is defined as the isotope ratio $^3\text{H}/^1\text{H} = 10^{-18}$. The typical precision of tritium measurement was equal to ± 0.3 TU.

Chemical and isotopic analyses were carried out at the Radio-Analysis and Environment Laboratory (LRAE) of the National Engineering School of Sfax (Tunisia) and the isotope hydrology laboratory at CNESTEN (Morocco). Carbon-14 and carbon-13 analyses (more than 60 samples) were performed by accelerator mass spectrometry (AMS) (Saito-Kokubu et al. 2015) and mass spectrometry, respectively, at the Isotopic Centre, University of Groningen (The Netherlands). The results of carbon-13 are expressed in δ values in permil units relative to the Pee Dee Belemnite standard (PDB). ^{14}C values are unnormalised and reported in percent modern carbon (pmc) (Cartwright et al. 2020).

Statistical analysis

In this investigation, multivariate statistical techniques such as correlation coefficients and principal component analysis (PCA) were performed on the chemical data (nine variables) of all the analyzed groundwater samples. Such methods have been widely used to investigate geochemical processes (Suk and Lee 1999; Lee et al. 2001; Kim et al. 2005; Shrestha and Kazama 2007; Matiatos et al. 2014; Trabelsi and Zouari 2019).

The preliminary step in the statistical analysis was calculating the Pearson correlation matrix (r). Generally, the parameters with $|r| > 0.7$ show a strong correlation, and $|r|$ between 0.5 and 0.7 indicates a moderate correlation (Manish et al. 2006).

Principal component analysis is a multivariate statistical method used to complement the limitations of graphical methods when inferring the dominant hydrogeochemical processes. The importance of the principal components is evaluated based on the score of the eigenvalues: principal components with the highest eigenvalues reflect the main geochemical processes governing the groundwater hydrochemistry (Suk and Lee 1999; Everitt and Hothorn 2011). In this study, PCA analysis was carried out with a minimum eigenvalue of 1. The varimax rotation method was used to extract the rotated component matrix with Kaiser normalization. Factor scores represented a measure of the magnitude of a principal component effect. A positive factor score was interpreted as a strong relevance of the indicated process in the study area. Negative values were interpreted as the area being negligibly affected by this process, whereas near zero indicates the moderate effect of the process.

The descriptive statistics of analytical results for all collected water samples were calculated using the Statistica 10 package. The multivariate statistical analysis was performed using the XLSTAT 2015 software.

Results

Chemical signature

Physical-chemical parameters

The temperature of the collected surface water samples varied from 14.8 to 28.4 °C (Table 2; Fig. 3), which could be linked to pumping rates and tubing length. The collected groundwater samples presented a more homogenous range of temperatures, except for some extreme values and outliers, which could be caused by artefacts during measurement.

These values should be viewed cautiously when computing thermodynamic equilibrium parameters. In general, mean water temperatures calculated for the different aquifers presented a limited variability, ranging from 29.3 to 31.8 °C except for some Quaternary groundwaters, characterized by lower temperatures, with a mean value of 27 °C.

Groundwater generally occurred as acidic to slightly alkaline, with pH values varying from 5 to 8.9, with typical values ranging from 7.1 to 7.7. However, pH values of the IC-CAM groundwaters presented more considerable variability, exceeding these limits with an SD equal to 1 (Fig. 3).

Groundwater salinity

Electrical conductivity (EC) and total dissolved solids (TDS) varied widely from one aquifer to another (Fig. 3). Surface water, and CTQ and basement groundwater samples presented the lowest and most homogenous levels of mineralization with

SDs ranging between 150 and 180 mg/L and mean TDS values equal to 167, 191, and 195 mg/L, respectively, except for some points showing a slight salinity increase. IC-CAM and Quaternary groundwaters displayed slightly higher salinity, with mean TDS values of 333.4 and 527.6 mg/L. In agreement with EC values, the CI aquifer was characterized by a substantial variability with an SD of 726 mg/L and higher mineralization, with TDS values ranging from 17 to 2,945 mg/L.

Chemical composition

The chemical composition of the collected groundwater samples showed marked variability within the same aquifer and from one aquifer to another (Fig. 4; Table 2). Calcium was present in high levels in the different water samples, ranging from 0.5 to 450 mg/L. The concentration of Na^+ varied over a wide range of values (from 0.6 to 550.9 mg/L). The highest concentrations were measured in the

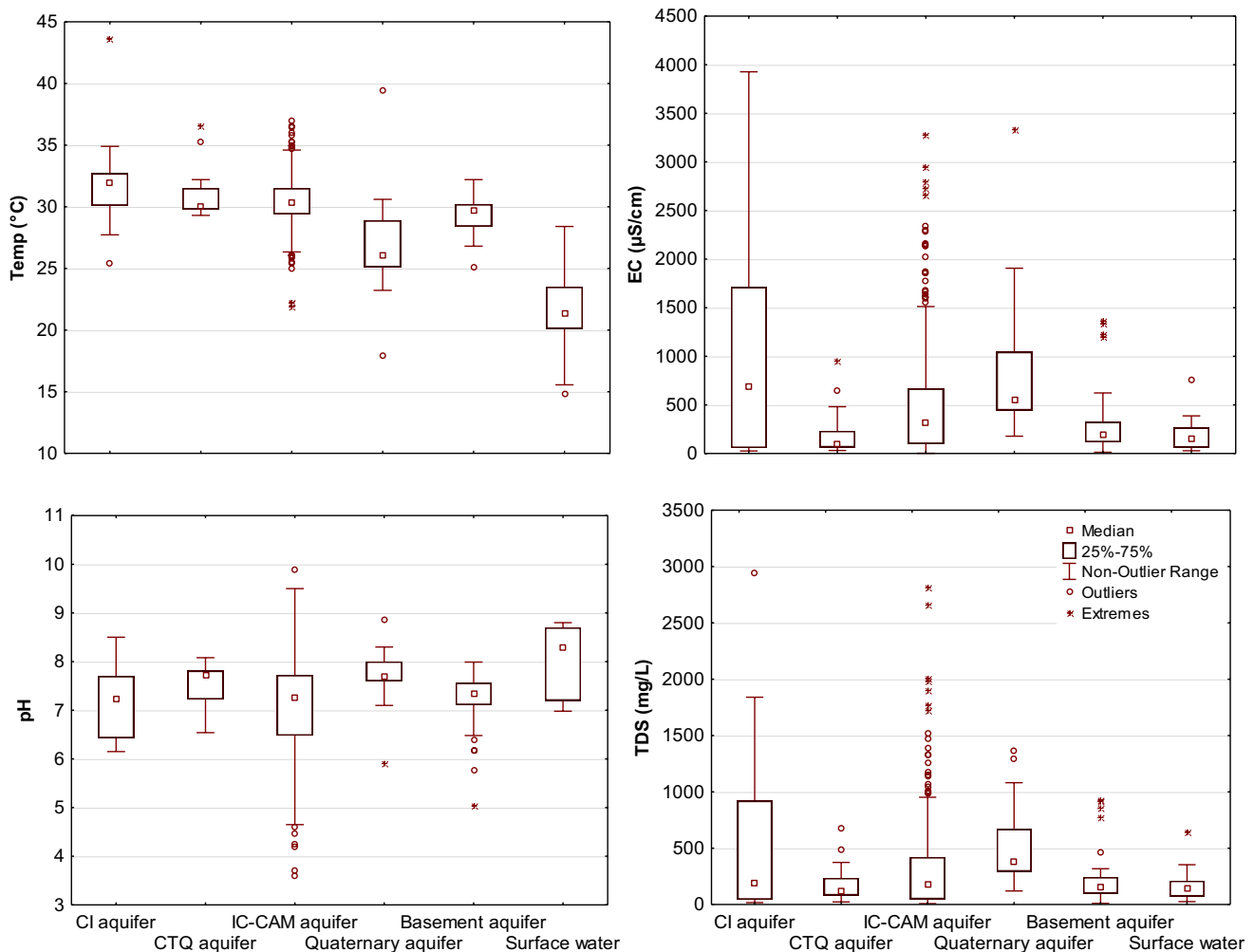


Fig. 3 Box plot of physical-chemical parameters—temperature, pH, electrical conductivity (EC), and total dissolved solids (TDS)—of the analyzed water samples. CI aquifer: Continental Intercalaire aquifer, CTQ: undifferentiated Continental Terminal-Quaternary Aquifer

CI and the IC-CAM groundwater samples. The contents of Mg^{2+} , ranging from the detection limit to 142.6 mg/L, probably result from the leaching of magnesium-bearing minerals from the mafic and ultramafic rocks. The study area is also mainly composed of biotite and dolerite in the Birrimian basement, and dolomite is found in several aquifer matrixes (IC-CAM, CI, and CTQ), hence, it may serve as a source of Mg^{2+} in these aquifers (Schlüter 2006). Potassium concentration varies from the detection limit up to 189.9 mg/L. The highest contents were recorded in the IC-CAM groundwaters.

HCO_3^- is the major anion present at the highest concentrations in surface water and groundwater samples collected from different lithologies, ranging from 2.1 to 884.5 mg/L. The concentrations of the dissolved sulfate ion ranged from the detection limit up to 1,341.3 mg/L. This anion showed the highest variability, with a high SD, particularly in the CTQ, the Quaternary, and the IC-CAM aquifers, where the sulfate content reaches the highest values. The concentrations of Cl^- , ranging from the detection limit up to 1,180.2 mg/L, were highly variable in the CI and the IC-CAM groundwaters, with an SD of ~280 mg/L. Nitrate varied from the detection limit up to 1,127.75 mg/L in groundwater. The highest concentrations were recorded in some sampling points of the basement and the IC-CAM aquifers.

The investigation of the leading chemical characteristics of the different aquifers according to the lithology (Fig. 4) revealed that the CTQ, the Quaternary, and the basement groundwaters presented the most homogenous composition compared to the CI and IC-CAM aquifers, which were characterized by a higher variation of the total mineralization.

Isotope hydrology

Isotope signature of rainwater

The isotopic contents ($\delta^{18}O$, δ^2H , and 3H) of monthly composite rain samples collected at Bamako station from May 1962 to October 1998 were used to define the isotopic composition of rainwater in the Taoudeni basin. The dataset used is available from the IAEA-GNIP database (IAEA/WMO 2020).

The calculated long-term mean weighted compositions were -4.5 and -28.4% vs SMOW, respectively, for $\delta^{18}O$ and δ^2H . These values agreed well with previous isotopic investigations in the Sahelian zone (Huneau et al. 2011; Gourcy et al. 2000; Taupin et al. 2000, 2002; Mathieu et al. 1993). The annual weighted mean values for rainfall were -4.2 and -23.5% respectively, for $\delta^{18}O$ and δ^2H at the Niamey station (Taupin et al. 2000, 2002). Similar isotopic values were found (-3.6% for $\delta^{18}O$ and -18.2% for

δ^2H) at the Bobo-Dioulasso station (Huneau et al. 2011) and (-3.7% ($\delta^{18}O$) and -23.8% (δ^2H)) at Barogo station (Mathieu et al. 1993). These studies confirmed that the origin of the air masses mainly controls the isotopic composition of rainwater in the Sahel region: the Guinean monsoon or easterly waves carried by the African Easterly Jet and Tropical Easterly Jet (Gourcy et al. 2000; Taupin et al. 2002). During humid years, the average isotopic composition of local precipitation is more negative because of the lower temperatures and reduced evapotranspiration (Gourcy et al. 2000).

The relationship between oxygen-18 and deuterium for the analyzed rainwater samples at Bamako showed that most of the samples aligned along the Global Meteoric Water Line (GMWL, Craig 1961; Fig. 5). These points determine a local meteoric water line (LMWL) defined by: $\delta^2H = 6.39 \delta^{18}O + 0.9$. A slope lower than 8 indicates the presence of fractionation processes leading to an enrichment in heavy isotopes. Evaporation has affected the isotopic composition of some rainwater samples. After removing the enriched points, the obtained LMWL was: $\delta^2H = 6.85 \delta^{18}O + 8.83$. This new line is closer to the GMWL and similar to that obtained by Gourcy et al. (2000), who worked on the same station from 1991 to 1998. ($\delta^2H = 8.1 (\pm 0.2) \delta^{18}O + 11.9 (\pm 2.1)$). Thus, during this study, the GMWL was used to characterize the isotopic signature of rainwater in the Taoudeni Basin.

Stable isotope ($\delta^{18}O/\delta^2H$) signature of groundwater and surface water

The isotope compositions of the collected water samples are presented in Fig. 6. The isotope measurements for surface waters showed high variability, ranging between -6.6 and $+29.7\%$ VSMOW, for $\delta^{18}O$ and between -41.9 and $+130.2\%$ VSMOW for δ^2H .

The distribution of the isotopic value was similar to the distribution found in precipitation. The most enriched samples were collected in December, and the isotopically most depleted in May. Such highly enriched values of $\delta^{18}O$ and δ^2H reflected a strong evaporation process affecting surface-water bodies.

The analyzed groundwater samples collected from the different aquifers were also highly variable with a standard deviation (SD) of 3.6% VSMOW for $\delta^{18}O$ values. Groundwater samples from the basement aquifer and the IC-CAM aquifer showed minimal variability in the $\delta^{18}O$ values with an SD of about 0.6 and 1% VSMOW, respectively. The CI groundwater presented the most depleted heavy isotopes content, with a mean value of -6.5% VSMOW for $\delta^{18}O$ and -48.0% VSMOW for δ^2H . The highest values of $\delta^{18}O$ and δ^2H are measured in the Quaternary and CTQ groundwaters, with $\delta^{18}O$ mean contents of 4.0 and -2.5% VSMOW, respectively (Table 2).

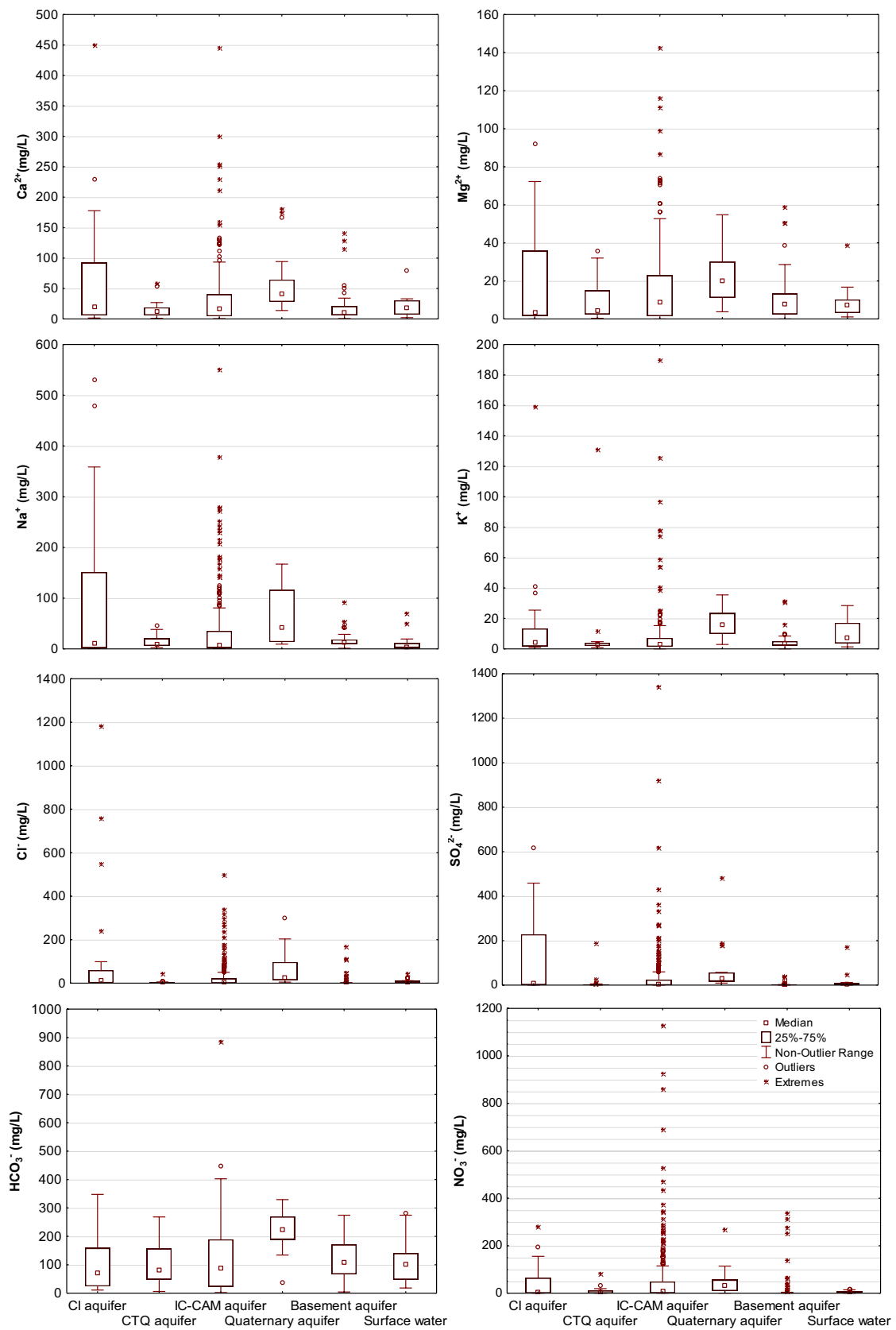


Fig. 4 Box plots of major chemical elements of the analyzed water samples. CI aquifer: Continental Intercalaire aquifer, CTQ: undifferentiated Continental Terminal-Quaternary Aquifer

Tritium contents in groundwater and surface waters

The monthly tritium contents of rainfall in the Bamako GNIP station showed that high contents, ranging between 64 and 1,111 TU, characterized the 1960s, whilst lower values were recorded in the 1970s with a mean value of 62 TU. The few measured tritium contents in the 1990s are much lower, with a mean value of 5 TU. Recently, rainfall samples collected within the regional project RAF7011 (2017) allowed for estimating the long-term annual tritium contents at ~4 TU, representing the atmospheric natural level in this area.

The measured tritium activities in the analyzed surface waters ranged from 1.7 to 6.7 TU (Table 2; Fig. 6). The most variable tritium contents were recorded in the IC-CAM, the CTQ, and the Quaternary aquifers where they could reach very significant values (>3 TU) with an SD exceeding 1.4 TU. Except for a few points, the measured tritium contents in the basement aquifer and CI groundwater samples were generally low, with mean values of 0.8 and 0.9 TU, respectively.

Carbon isotopes in groundwater

Carbon isotopes ($\delta^{13}\text{C}/^{14}\text{C}$) were analyzed in various aquifer systems to obtain further information on the time scales of groundwater residence time, as well as the geochemical reactions within the carbon system (Clark and Fritz 1997).

The measured ^{14}C activities and $\delta^{13}\text{C}$ values of the total dissolved inorganic carbon (TDIC) covered a wide range of values, reflecting the presence of several sources of dissolved carbon in groundwater and different transit times.

The most variable ^{14}C activities were measured in the CI groundwaters, ranging from 1.7 to 86.1 pmc. This range revealed relatively recent recharge in addition to palaeo-recharged waters. The lowest radiocarbon activities were measured in the northern part of the basin, in the Algerian part, where the aquifer is highly confined. The highest carbon-14 activities were found in water samples collected from the Quaternary aquifers, confirming the existence of recent, abundant recharge. IC-CAM and basement groundwater samples showed carbon-14 activities ranging from 15.6 to 116.8 pmc and 51.8 to 97.6 pmc, respectively, pointing to a binary mixing of old and modern groundwater.

The carbon-13 contents varied widely also within the studied aquifers (Table 2; Fig. 6) with an SD of ~3‰ VPDB. CI groundwater presented relatively enriched contents in $\delta^{13}\text{C}$, with a mean value of -7.7‰ VPDB. However, the most depleted carbon isotope contents were measured in the basement aquifer, with a mean value of -13.3‰ VPDB, indicating that the primary source of inorganic dissolved carbon in this system is probably soil gas CO_2 . This is in agreement with previous isotopic investigations performed on the Sahel region which prove that carbon-13 contents of

soil gaseous CO_2 range from -19 to -13‰ versus PDB (Le Gal La Salle 2001; Huneau et al. 2011). The IC-CAM and the Quaternary groundwater samples presented a mean $\delta^{13}\text{C}$ content of about -10.5‰ VPDB, suggesting a geochemical interaction between biogenic CO_2 and the carbonate matrix.

Discussion

Hydrochemical investigation

Regional assessment: water types

Water types are often defined based on the chemical composition of the investigated water samples. This classification correlates the contents of major ions in groundwater with the geological and hydrogeological settings of the studied aquifer systems. A graphical representation of the relative concentrations of major ions using a Piper diagram (Piper 1944) (Fig. 7) displayed different water types: Na-K- HCO_3 , Ca-Mg- HCO_3 , Ca-Mg-Cl- NO_3 - SO_4 , Na-K-Cl- SO_4 , and Ca-Mg-Na-K- SO_4 . In the present study, the chemical compositions of the collected samples were classified according to five water types: water group 1 (Na- HCO_3), group 2 (Ca-Mg- HCO_3), group 3 (Ca-Mg-Cl- SO_4), group 4 (Na-Cl- SO_4) and group 5 (Ca-Mg-Na- SO_4) (Table 3).

Water group 2 dominated in surface water samples and almost all the aquifers, regardless of their hydrogeological settings (Table 3). From the Piper plot, it was clear that there was an excess of alkaline earth ions (Ca^{2+} , Mg^{2+}) when compared to alkalis (Na^+ , K^+). The presence of these elements in groundwater could result from water interactions and weathering of major rock types resulting in the release of Ca^{2+} , Mg^{2+} , and HCO_3^- in groundwater (Adithya et al. 2016; Huneau et al. 2011). This water type could reflect the natural recharge of groundwater and short residence time (Prasanna et al. 2010; Trabelsi et al. 2020; Thivya et al. 2013).

Water group 1 dominates in about 20% of the collected samples of the Basement and CTQ aquifers, 13% of the Quaternary groundwater samples, 8% of the IC-CAM, and 6% of surface-water samples. The Na^+ and K^+ may primarily enter the groundwater in the study area through the incongruent dissolution of feldspars, plagioclases, and biotite or due to anthropogenic activities (deicing, fertilizer and wastewaters). In addition, cation exchange reactions of Ca^{2+} and Mg^{2+} could lead to the release of Na^+ and K^+ into groundwater (Huneau et al. 2011; Trabelsi et al. 2020).

Water group 3 included about 30% of the IC-CAM and Quaternary groundwater samples and 19% of the CI groundwater. It was not detected in the CTQ aquifer and did not exceed 7% of the basement and surface-water samples.

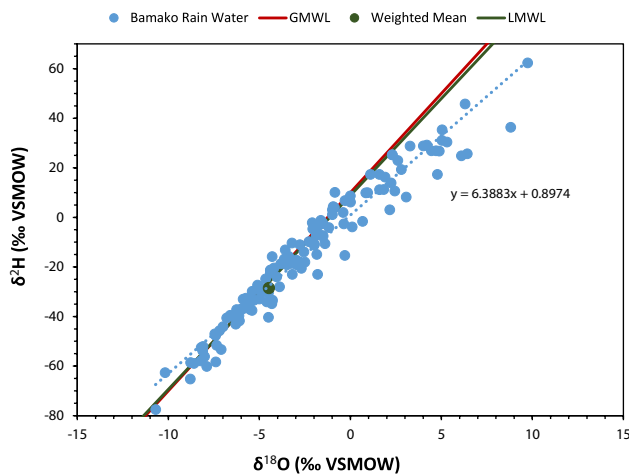


Fig. 5 Oxygen-18 vs deuterium ($\delta^2\text{H}$) diagram of rainfall in Bamako station showing the weighted mean annual composition of rain. The red line is the global meteoric water line (GMWL). The green line represents the local meteoric water line (LMWL) obtained after removing the isotopically enriched points. The dashed blue line represents the LMWL with all data. These lines were calculated using the least-squares regression method

The presence of chloride and sulfate in these groundwater samples indicated the existence of an advanced stage of water–rock interaction through the dissolution of sulfate minerals (gypsum and anhydrite). However, the increase in the nitrate ion in some samples reflected the effect of anthropogenic contamination.

Water group 4 was identified in about 15% of the CI groundwater samples, 6% of the CTQ and the IC-CAM aquifer, and 4% of the Quaternary. Groundwaters belonging to this water type were enriched in sodium and chloride, except for a few points that presented a slight increase in the contents of the sulfate ion. This chemical composition probably derives from water–rock interaction processes (e.g., halite or silicate weathering; Mukherjee et al. 2009). Similarly, Na^+ may be derived from cation exchange reactions through clay minerals.

The last water type, group 5, is only found in 12 and 6% of the CI and the CTQ groundwater samples. The main characteristic of this group was the dominance of sulfate in groundwater mineralization. Calcium, magnesium, and sodium were present in the same proportions.

At the regional scale, the dominance of water groups 1 and 2 suggested the existence of the same geochemical processes affecting several aquifers with diverse stratigraphic and geological layers. In addition, different water types could be found in a single aquifer, indicating the heterogeneity of the geochemical processes within a unique hydrogeological system. The scattering of sample points within the same aquifer, and concerning all the other aquifers, is seen in the Piper plot in Fig. 7 (Table 3), highlighting the complex

petrographical nature of the aquifer formations, producing geochemical variances from one borehole to another. Thus, individualized groundwater flow paths are difficult to delineate using only the chemical composition within the different aquifer layers.

Statistical identification of geochemical processes

Pearson's correlation matrix, presented in Table 4, provides further insight into the possible ion sources in groundwater in the Taoudeni basin. A strong correlation was found between the major chemical ions (Ca , Mg , Na , Cl , SO_4 , NO_3 and HCO_3) and TDS with correlation coefficients exceeding 0.6 (Table 4), which clearly indicates the contribution of these ions to the overall mineralization. A significant positive correlation was observed between TDS and HCO_3 , Ca , Mg and Na (Table 4), meaning these ions may originate from carbonate or silicate dissolution. A notable correlation exists between NO_3 , Ca , Mg and Cl concentrations, which may mean that groundwater is highly affected by anthropogenic contamination.

The processes controlling the chemical quality of groundwater in the Taoudeni basin can be identified by using PCA. This analysis included the variable loadings, eigenvalues and variance explained by each factor (Table 5). Two factors were extracted, which described 73.1% of the total data variance with eigenvalues greater than 1. The first principal component explained the largest variability (60%) in the data and represented the most critical process or combination of processes controlling water chemistry (Yidana and Yidana 2010). Although the statistical study was performed on groundwater samples of different lithological and stratigraphic characteristics, the PCA analysis revealed the dominance of a single component (F1) that governs the chemical quality of these waters. This principle component (F1) showed a strong positive correlation with Cl^- , Mg^{2+} , Na^+ , TDS, and Ca^{2+} and moderate loading for HCO_3^- and SO_4^{2-} (Fig. 8). This component probably represented a series of processes, including silicate weathering and mineral dissolution under the water–rock interaction, which increase groundwater salinity.

Principle component F2 explained 12.8% of the total variance with a positive correlation with K^+ (0.6) and NO_3^- (0.5), which reflected nitrate contamination (Zhou et al. 2007) and highlighted the effect of the use of chemical fertilizers, in particular, potassium-based composite. The opposite position of Na^+ compared to Ca^{2+} and Mg^{2+} indicated the fixation and release of this cation through ion exchange reactions supported by clay minerals within the aquifer matrix.

The factor scores of the sample reflected the strength of the geochemical process represented by the principal

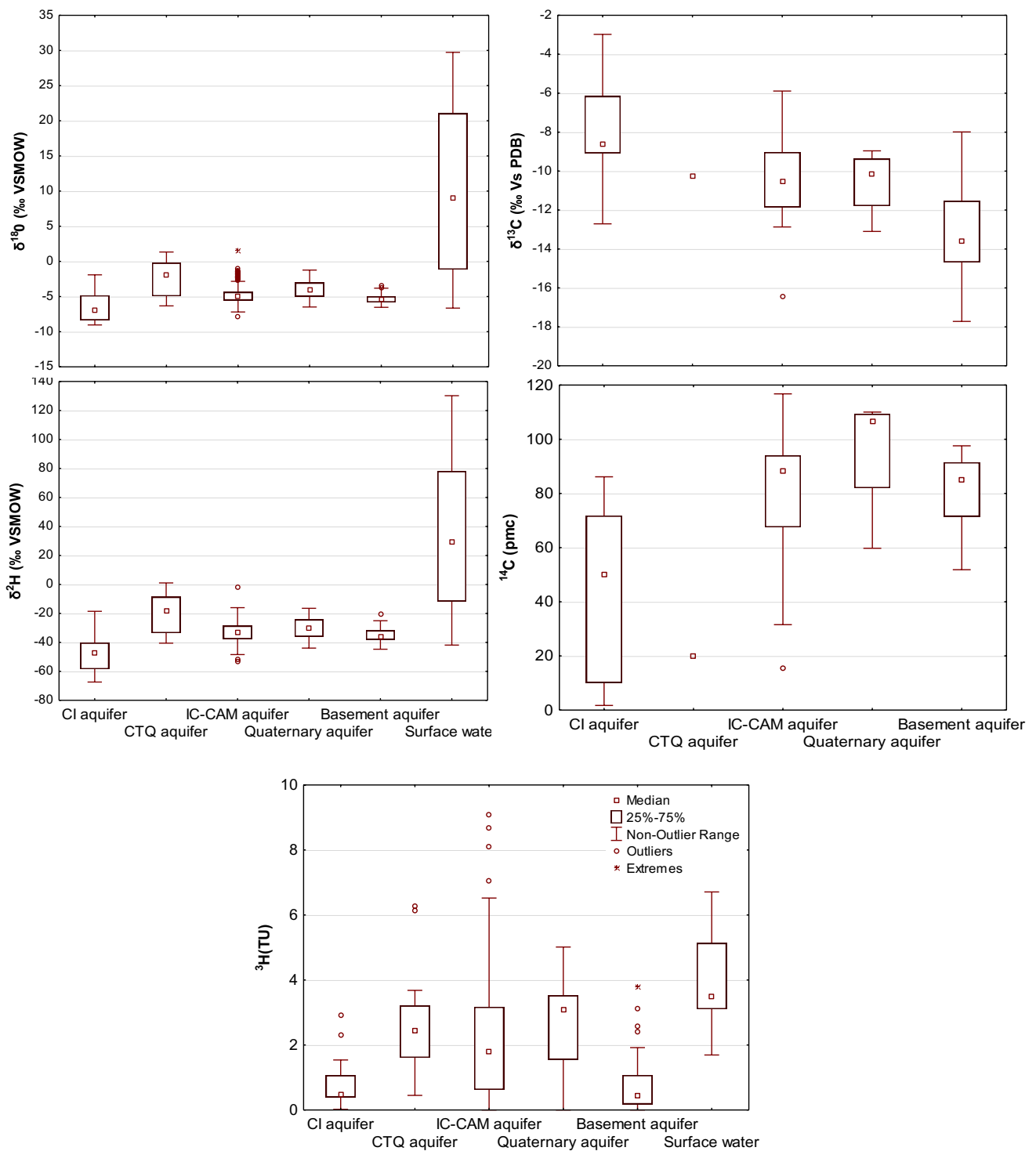


Fig. 6 Box plots displaying the isotopic parameters of the analyzed groundwater samples in the Taoudeni basin in the main aquifers. $\delta^{18}\text{O}$ and $\delta^2\text{H}$ contents of groundwater samples are plotted again relative to the GMWL in Fig. 12

factors and can be considered a measure of the chemical impact of the geochemical processes on groundwater in the study area. In the scatter plot in Fig. 9, about 30% of groundwater samples show positive factor scores F1 and

F2. This suggests that these processes have moderate effects on the hydrochemical data. The higher impact of factor F1 occurred in the groundwater of IC-CAM, CI and some CTQ samples, which belong to groups 3, 4 and 5. Most of the

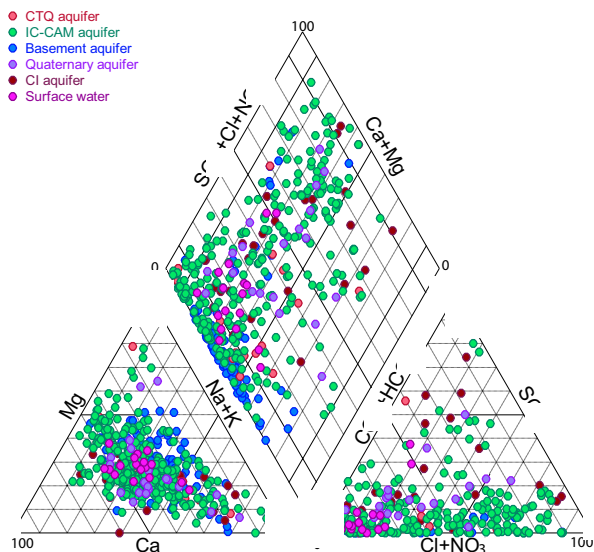


Fig. 7 Piper diagram showing the main water types within the different aquifers of the Taoudeni Basin

basement groundwaters showed a negative score for factor F1, which could be explained by the low salinity of these samples reflecting limited water–rock interaction. However, the high factor scores F1 and F2 observed mainly in IC-CAM revealed that the increase of groundwater salinity was accompanied by the increase of Mg, Ca and NO_3

concentrations, reflecting an anthropogenic impact. Furthermore, groundwater samples of IC-CAM and CI groundwater belonging to groups 1 and 4 evolve towards the pole of SO_4 , HCO_3 and Na (positive score for F1 and a negative score for F2). This tendency traduces that the increase of groundwater salinity in these samples is mainly related to an advanced degree of water–rock reaction such as intense silicate weathering and cation exchange processes.

Major geochemical processes

In the studied aquifer systems, the chemical composition of groundwater is mainly controlled by the interaction of infiltrating waters with the rock matrix through various processes. It is tough to differentiate these reactions based on a mass solute balance. Unfortunately, the detailed mineralogical compositions of the various aquifer formations were not available in the Taoudeni basin. There are some general geological data regarding the dominant rock types at the level of the whole basin. Silicates (quartz, feldspar, micas, biotite, etc.), carbonates (calcite and dolomite), gravel particles, sandstones and clay minerals form the main aquifer formations (Shields et al. 2007).

Several diagrams were employed to investigate the relative contributions of the major weathering/dissolution reactions (silicate, carbonate, and evaporite) to the

Table 3 Distribution of the water type groups (%) and the dominant water type group for each aquifer

Group	Water Type	Basement	CI aquifer	CTQ aquifer	IC-CAM aquifer	Quaternary	Surface water
1	Na–K– HCO_3	21	4	19	8	13	6
2	Mg–Ca– HCO_3	72	50	69	56	52	88
3	Ca–Mg–Cl– SO_4	7	19	0	29	30	6
4	Na–Cl– SO_4	0	15	6	6	4	0
5	Ca–Mg– NaSO_4	0	12	6	1	0	0

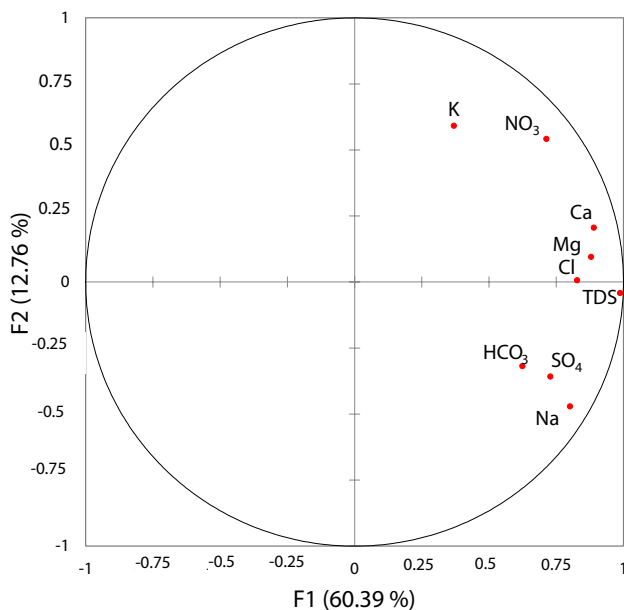
Table 4 Pearson's correlation matrix of chemical elements calculated for all collected samples

Variables	TDS	Ca	Mg	Na	K	Cl	SO_4	NO_3	HCO_3
TDS	1								
Ca	0.87	1							
Mg	0.85	0.77	1						
Na	0.82	0.53	0.55	1					
K	0.34	0.30	0.29	0.12	1				
Cl	0.80	0.75	0.69	0.67	0.22	1			
SO_4	0.76	0.65	0.59	0.71	0.13	0.49	1		
NO_3	0.69	0.77	0.68	0.35	0.36	0.56	0.25	1	
HCO_3	0.61	0.39	0.56	0.67	0.24	0.39	0.30	0.23	1

Table 5 Classification of the principal components according to the obtained eigenvalues

Parameter	F1	F2	F3	F4	F5	F6	F7	F8	F9
Eigenvalue	5.44	1.15	0.89	0.64	0.40	0.28	0.18	0.02	0.00
Explained variance (%)	60.39	12.76	9.88	7.10	4.48	3.15	1.96	0.24	0.04
% cumulative variance	60.39	73.15	83.03	90.14	94.62	97.77	99.73	99.96	100.00

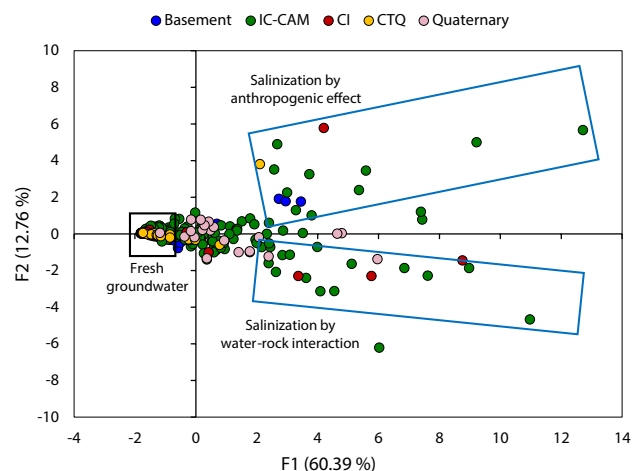
ion concentrations in groundwater (Figs. 10 and 11). The influence of carbonate dissolution and silicate weathering was evaluated using bivariate mixing diagrams of Na^+ -normalized Mg^{2+} versus Na^+ -normalized Ca^{2+} and Na^+ -normalized HCO_3^- versus Na^+ -normalized Ca^{2+} (Fig. 10). In these diagrams, the approximate compositions of three end-members, namely the carbonate dissolution, silicate weathering and evaporite dissolution, were represented according to the model suggested by Gaillardet et al. 1999. These bivariate plots showed that basement groundwater and most IC-CAM samples that belong to group 1 were within or very close to the global average silicate-weathering domain, which indicates a significant contribution of silicate mineral weathering dissolution to the groundwater chemistry of these samples. This process occurs at the shallow part of the aquifers (open system) in the presence of oxygen at a relatively lower temperature. Some samples of IC-CAM were close to the carbonate end member, indicating the dominance of the dolomite and/or calcite dissolution on the chemical quality in these few groundwater samples. However, most groundwater samples of different aquifers are plotted in the transitional area between the carbonate end member and the silicate-weathering zone, illustrating a combined effect of carbonate and silicate dissolution.

**Fig. 8** Plots of PCA components of the chemical parameters of groundwater in the Taoudeni aquifer system

The bivariate diagram molar ratio $\text{Mg}^{2+}/\text{Ca}^{2+}$ versus Mg^{2+} presented in Fig. 11a showed that the dolomite dissolution was emphasized over the calcite dissolution, which contributed to the increase of solutes (Ca^{2+} , Mg^{2+} , HCO_3^-) concentrations in the studied groundwater. In some groundwater samples, the $\text{Mg}^{2+}/\text{Ca}^{2+}$ molar ratio was low, suggesting the influence of calcite dissolution. However, most of the water samples of IC-CAM (group 3) are, in contrast, slightly enriched in Ca^{2+} and Mg^{2+} relative to bicarbonate, reflecting that the carbonate mineral dissolution is minor and there are other sources of Ca^{2+} and Mg^{2+} ions in these groundwaters. Significant correlations were also observed between $\text{Ca}^{2+} + \text{Mg}^{2+}$ ions and NO_3^- concentrations in the binary diagram of Fig. 11c, which indicates that dissolved ions probably originated from anthropogenic sources such as fertilizers and/or wastewater.

Additionally, Fig. 10 shows that some groundwater samples tended to fall between the silicate weathering end member and the evaporite dissolution, which suggested that silicate weathering is prevailing and that evaporite dissolution has a negligible influence on solutes in groundwater.

Besides the carbonate dissolution and silicate weathering processes, cation exchange reactions can affect groundwater quality. To evaluate the influence of cation exchange processes in the Taoudeni groundwater, an equivalent bivariate plot of corrected bivalent versus corrected monovalent cations is presented in Fig. 11c. In this diagram, concentrations of Ca^{2+} and Mg^{2+} that may have been involved in

**Fig. 9** Scatterplot of the first two principal component (F1 and F2)

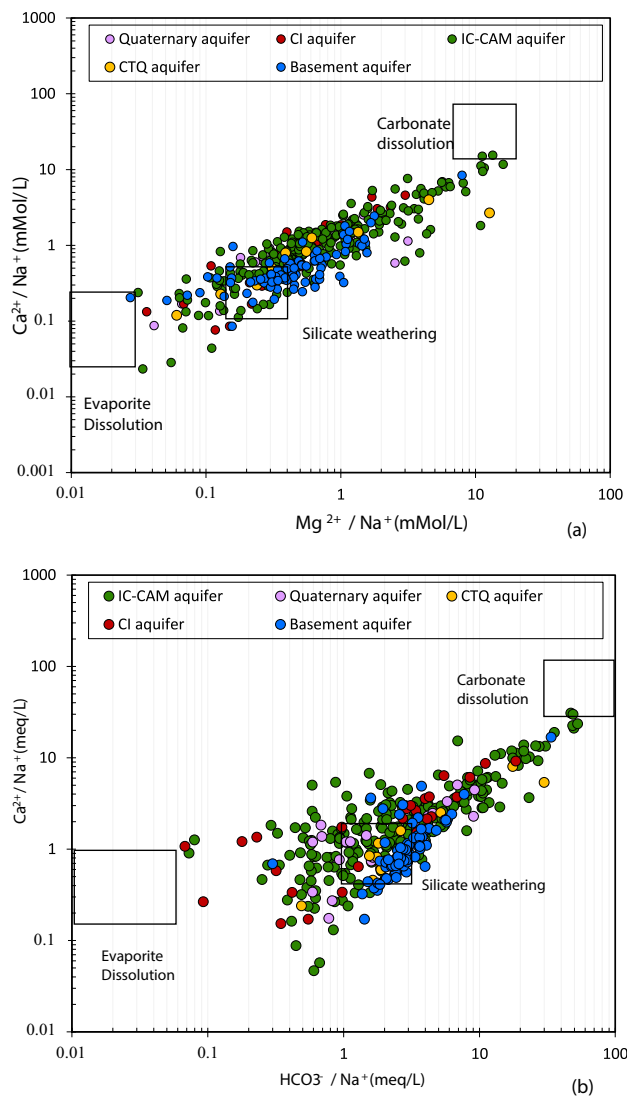


Fig. 10 Molar ratio bivariate plots of **a** Na-normalized Ca and Mg, and **b** Na-normalized Ca and HCO_3^- . The three rectangles represent the approximate compositions of the three primary source end members (evaporites, silicates, and carbonates) without mixing

exchange reactions were corrected by subtracting equivalent concentrations of associated anions (HCO_3^- and SO_4^{2-}) that would be released by carbonate and gypsum dissolution and or/silicate weathering (Mukherjee et al. 2009). Similarly, monovalent cations ($\text{Na}^+ + \text{K}^+$) that may be derived from the aquifer matrix would be balanced by equivalent concentrations of Cl^- (McLean and Jankowski 2000). The slope of -1 in this bivariate plot indicates that active cation exchange was taking place within the clay mineral of the aquifer (Jankowski et al. 1998). Figure 11c shows that most of the water samples are plotted close to the origin, indicating that cation exchange does not influence the groundwater chemistry, except for some points of the CI and IC-CAM aquifer, which are close to a line with a slope of -1 . This observation

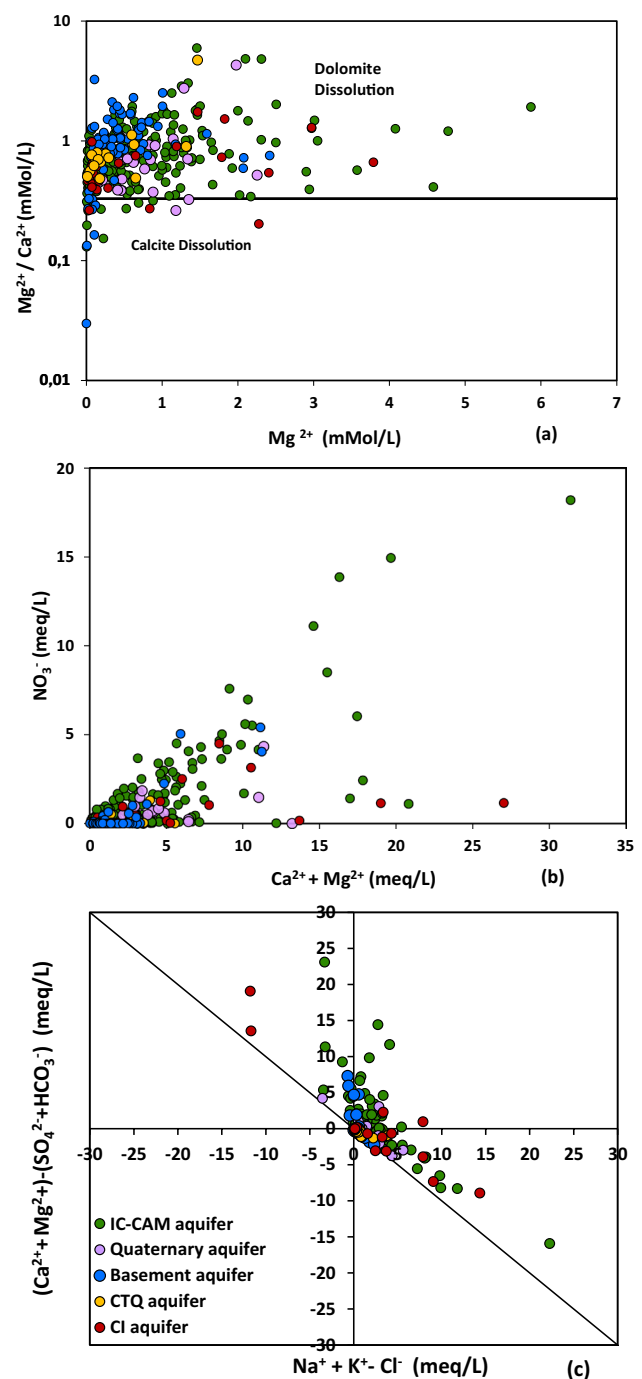


Fig. 11 Diagrams of **a** Molar ratio $\text{Mg}^{2+}/\text{Ca}^{2+}$ versus Mg^{2+} , **b** NO_3^- versus $\text{Ca}^{2+} + \text{Mg}^{2+}$, **c** Bivariate plot of $(\text{Ca}^{2+} + \text{Mg}^{2+}) - (\text{HCO}_3^- + \text{SO}_4^{2-})$ against $(\text{Na}^+ + \text{K}^+ - \text{Cl}^-)$ to indicate the cation exchange reactions in collected groundwater samples in the Taoudeni basin

indicates that cation exchange dramatically influences the water chemistry of these samples.

The groundwater residence time, as well as the hydrological conditions, influence the cation exchange processes (Guo and Wang 2004; Rajmohan and Elango 2004; Huneau et al. 2011). The magnitude of the cation exchange processes in

the CI aquifer and some parts of the IC-CAM aquifers is enhanced by relatively long groundwater residence times.

The isotopic investigation

Groundwater origin

The stable isotope composition of groundwater has been extensively used for identifying recharge processes, determining evaporation effects and investigating the interactions between surface and groundwaters (Clark and Fritz 1997; Trabelsi et al. 2020). The measured values of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ of the Taoudeni groundwater varied from -67 to 1‰ for $\delta^2\text{H}$ and -9 to 1.6‰ for $\delta^{18}\text{O}$. Most groundwater samples of the different aquifers are situated close to the GMWL in Fig. 12. Depleted $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values were measured in some water samples belonging to the confined part of CI aquifer in Algeria. Such isotopically depleted values were not observed elsewhere in the Taoudeni basin. They are interpreted as paleowaters, confirmed by their lowest radiocarbon activities and zero tritium contents (Fig. 13).

Groundwater samples of the basement, most IC-CAM samples and some points of the Quaternary and CTQ aquifers are located close to the GMWL. The stable isotope signature of these water samples, which is in the range of present-day regional and global meteoric water, is linked to a limited evaporation effect due to direct and rapid rainwater infiltration.

Surface-water samples present an enriched composition in $\delta^{18}\text{O}$ and $\delta^2\text{H}$ and plot along an evaporation line in Fig. 12. This observation reveals that the concentrations of ions in surface water, particularly in Mauritania, increased under conditions of strong evaporation, enhanced by climate aridity. It is noticeable that some points of the IC-CAM, CTQ and Quaternary aquifers were isotopically enriched and plot on the same line as surface waters, pointing to the

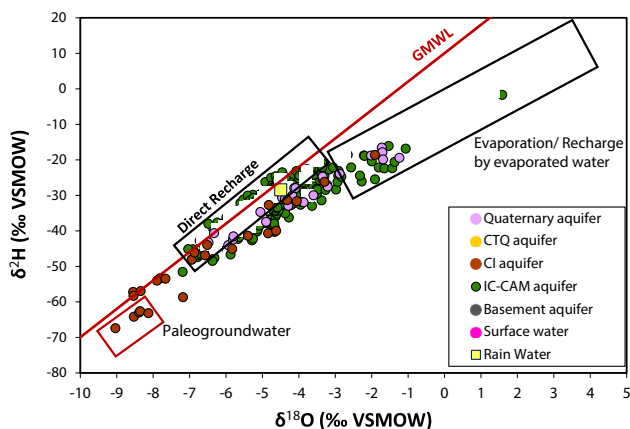


Fig. 12 Plot of $\delta^{18}\text{O}$ versus $\delta^2\text{H}$ of the different aquifers of the Taoudeni Basin compared to the GMWL

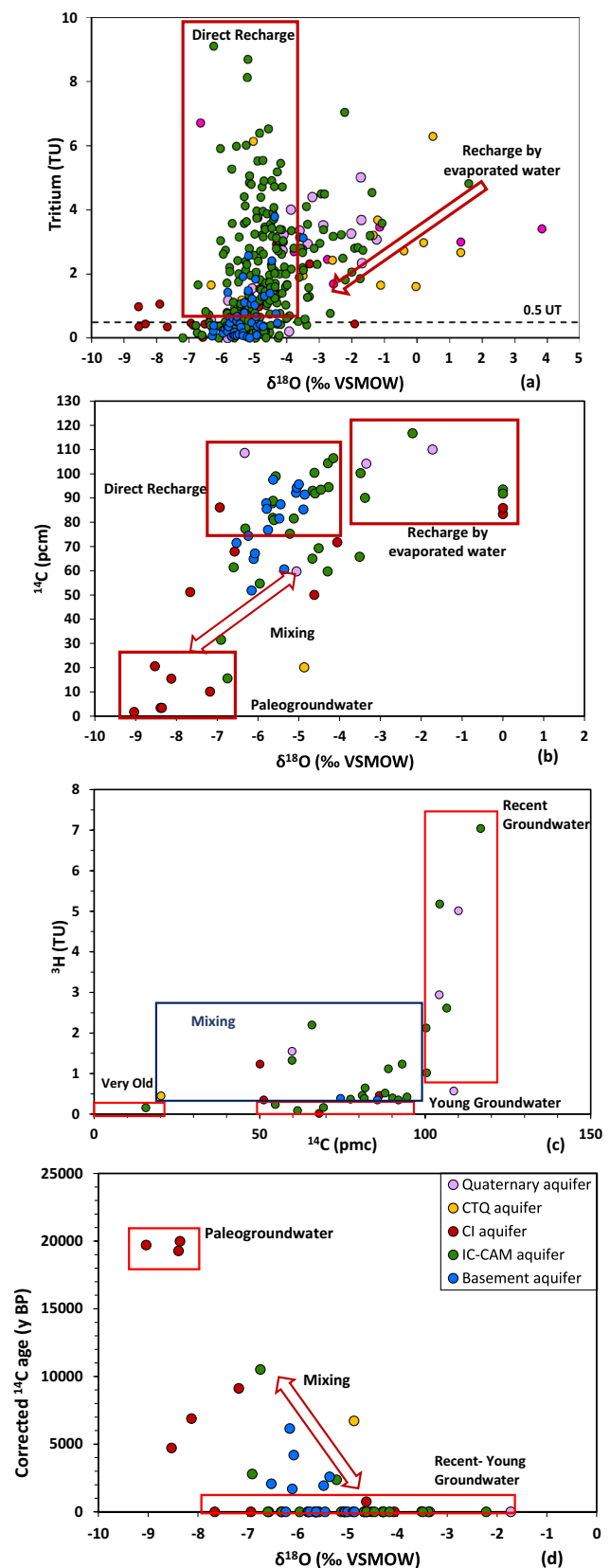


Fig. 13 Plots **a** $\delta^{18}\text{O}$ vs tritium, **b** $\delta^{18}\text{O}$ vs carbon-14 activities, **c** tritium vs carbon-14 activities, **d** corrected ^{14}C age vs $\delta^{18}\text{O}$. See text for details

existence of a hydraulic communication between surface water and groundwaters.

Recharge processes

^3H concentrations and ^{14}C activities were used to classify the groundwater samples into four age categories (recent, young, mixed and very old) (Fig. 13d). Recently recharged groundwater is characterized by measurable tritium contents (>0.5 TU) and high ^{14}C activities. These groundwaters are not older than some tens of years.

This group includes most points of the basement, IC-CAM, Quaternary and CTQ aquifers. These isotope contents indicate a recent recharge that occurred over 10–20 years, with tritium contents ranging between 1 to 4 TU. Also, there are some water samples of the IC-CAM and CTQ which present tritium contents higher than 5 TU revealing a recharge during the period of tritium peak (1955–1980), with content >5 –6 TU. The occurrence of recent groundwater that was recharged by precipitation of the 1960s is also supported by high carbon-14 activities exceeding 100 pcm, particularly in the IC-CAM aquifer.

This recent recharge could be direct by fast infiltration of rainwater as in the case of the majority of IC-CAM and the basement groundwater samples or indirectly through the infiltration of evaporated surface water as suggested by the enriched stable isotopes composition of Quaternary and CTQ groundwater, particularly in Mauritania (Fig. 13).

Groundwater quality of this group is marked by the dominance of Ca–Mg– HCO_3 type with low salinity, which reflects an earlier stage of chemical development and the contribution of meteoric water and surface water to the recent recharge of the aquifer.

Young groundwater includes water samples with significant ^{14}C activities and tritium content lower than 0.5 TU, which implies a contribution of older water (before 1950). This group of water may be hundreds of years old. It comprises some samples of IC-CAM and CI aquifers located mainly in the eastern part of Mauritania.

Very old water is characterized by groundwater samples with ^{14}C activities less than 20 pcm and with low tritium values. This groundwater, also called fossil, is several tens of thousands of years old. This is also the case of CI groundwater exploited in the southern part of Algeria and some IC-CAM groundwater. The long residence time of these groundwaters has enhanced geochemical reactions with the aquifer matrix, such as cation exchange processes.

Groundwater samples presenting as relatively high in ^3H and lower in ^{14}C than recent water are likely young and old water mixtures. This mixing between old and recently recharged groundwaters characterizes the regional sedimentary aquifer as the CI and IC-CAM aquifers, as highlighted

by the binary isotopes diagram (Figs. 12 and 13). The evolution from an open to a closed system is accompanied by a change of water type from Mg–Ca– HCO_3 and Na–K– HCO_3 (water groups 2 and 1) to Ca–Mg–Na– SO_4 (water group 5). Thus, the geochemical processes evolved from minor dissolution of dolomite and calcite and silicate weathering to a more advanced interaction with the aquifer matrix as a cation exchange reaction.

In the present investigation, the geochemical correction model of Fontes and Garnier (1979) using measured variables such as alkalinity, pH, temperature and $\delta^{13}\text{C}$ values of dissolved inorganic carbon (Fontes and Garnier 1979) is applied to convert the measured carbon-14 activities into groundwater ages. This model is well adapted to the geochemical reactions experienced by groundwater within the aquifer (Huneau et al. 2011). The obtained radiocarbon ages range from modern recharge to the late Pleistocene (Fig. 13d). Mixing of groundwaters of various ages is observed in the IC-CAM aquifer, in the basement aquifers, and in the CI present, where groundwater ages ranged from 1.6 to 10.5 ka BP. The confined part of the CI aquifer in Algeria presents groundwater ages of around 20 ka BP. The depleted stable isotope signature of these geochemically evolved groundwaters most likely indicates the presence of fossil waters, recharged during cooler, more humid phases before the Last Glacial Maximum (LGM). The late Pleistocene was characterized by an increased frequency of heavy rain events (Beyerle et al. 2003). The absence of groundwater samples with ages ranging from 10.5 to 20 ka BP indicates dry conditions in the studied region during the LGM with no significant recharge.

This finding is also in agreement with the previous results obtained in the Burkinian part by Huneau et al. (2011), in northern Mali by Fontes et al. (1993) and in Niger by Andrews et al. (1994) and Beyerle et al. (2003).

Nitrate contamination

The nitrate concentration vs $\delta^{18}\text{O}$ binary diagram (Fig. 14) revealed that samples representing the impact of anthropogenic processes (with $\text{NO}_3^- > 50$ mg/L) belonged to the IC-CAM, Quaternary and basement, and ranged from depleted to enriched isotopic contents, resulting in a complex geochemical process involving evaporated water bodies. One of the significant sources of nitrate pollution is contamination by domestic sewage, and the use of fertilizers and pesticides in the Taoudeni basin. High NO_3^- concentrations characterize samples that were mainly collected in rural areas, where a great deal of domestic sewage, agricultural fertilizer and pesticides had been discharged into the nearby ground or ditches, infiltrating into the local groundwater bodies.

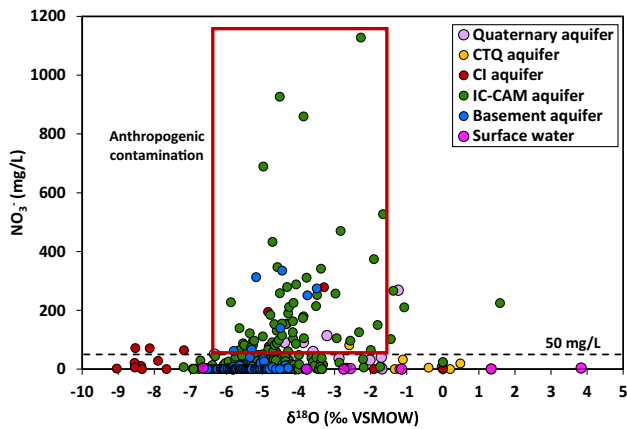


Fig. 14 $\delta^{18}\text{O}$ versus nitrate for the different aquifers of the Taoudeni Basin

Furthermore, the occurrence of high nitrate concentrations in the CI groundwaters in the Algeria Saharan region could be explained by natural processes. Nitrate naturally exists in the Sahelian region's soil, characterized by particular vegetation types, like acacia, a nitrogen-fixing tree, as well as the presence of termites, which are also known to produce nitrogen (Ngugi and Brune 2012).

Conclusions

This investigation illustrates how combining isotopic and hydrochemical tools and multivariate statistical analysis may improve the knowledge of groundwater geochemical processes that control groundwater quality in a complex hydrogeological and geological setting such as the Taoudeni aquifer system. In this shared basin, groundwater resources are extracted from several lithological and stratigraphic formations (basement, consolidated sediments, and alluvium). The assessment and management of groundwater resources require a solid understanding of the hydrogeological and hydrogeochemical properties of the aquifers. PCA revealed the dominance of two principal components or sources responsible for water quality variations in the Taoudeni aquifers. The main geochemical processes controlling groundwater quality are related to water–rock interaction, such as mineral dissolution and silicate weathering. The anthropogenic effect was identified as the second major factor responsible for water quality deterioration, causing a marked increase in nitrate contents and chloride, magnesium, and calcium concentrations.

The identified hydrochemical processes indicated slow hydrodynamics, the occurrence of carbonate dissolution and silicate weathering in the recharge zone, and the occurrence of cation exchange in the most confined part of the shared aquifer. The less mineralized water samples

exhibited a trend toward the HCO_3^- , Ca, and Mg components. The dissolution of calcite and particularly dolomite controlled the presence of these ions in groundwater. Silicate weathering was also identified to occur in several aquifers, such as the basement and CTQ aquifers. The most mineralized groundwaters were mainly linked to anthropogenic activities (agricultural and domestic practices), where local nitrate contamination was detected in the different aquifers.

The stable isotope composition of most groundwater samples indicated a clear meteoric origin. The importance of recent and direct recharge was prominent in the basement, IC-CAM, CTQ, and Quaternary aquifers, where tritium contents were significant and radiocarbon activities were high. However, depleted stable isotopic contents and low ^{14}C activities characterized the groundwaters of CI aquifers in the northern part of the basin, where an arid climate prevails. These isotopic contents indicated a palaeoclimatic recharge with different atmospheric circulation compared with current climatic conditions.

In summary, groundwater is the primary source of reliable freshwater in Taoudeni basin, and is used for irrigation, domestic, and industrial water supplies. Unfortunately, several issues linked to water scarcity and quality degradation threaten this precious resource, which needs to be adequately managed and protected as a high-quality resource and as part of the integrated plans for the basin's future water supplies. For this reason, further investigations should be performed to evaluate the effects of mining, industrial, and agricultural activities and the impact of urbanization on the groundwater resources in the Taoudeni basin.

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Declarations

Conflicts of interest No potential conflict of interest was reported by the authors

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