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Isotope investigation of shallow aquifers in Chott Djerid, Southwestern Tunisia

Indagine sugli isotopi delle falde acquifere poco profonde a Chott Djerid, Tunisia sudoccidentale

Zohra KRAIEM[a] ✉ , Kamel ZOUARI[a], Najiba CHKIR[a,b], Aissa HLEIMI[c]

[a] Radio-Analysis and Environment Laboratory, the National Engineering School, University of Sfax, Sfax, Tunisia
e-mail ✉ : *zohra.kraiem@enis.tn*
[b] Department of Geography, Faculty of Letters and Humanities, P. Box 1168-3002 Sfax, Tunisia
[c] Water resources directory, Ministry of Agriculture, Water Resources and Fishing, Tunisia

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Correspondence to:
Zohra Kraiem ✉ :
zohra.kraiem@enis.tn

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Riassunto

Le falde acquifere sono una risorsa cruciale per vari scopi, soprattutto nell' arida Tunisia meridionale. La valutazione delle proprietà richiede una comprensione approfondita dei principali processi di mineralizzazione. Pertanto, è stata effettuata una caratterizzazione completa delle falde acquifere superficiali nell'area di Chott Djerid utilizzando metodi isotopici basati su ossigeno-18, deuterio, trizio, carbonio-14 e carbonio-13 su 46 campioni di acqua. I risultati ottenuti mostrano che sia l'ossigeno-18 che il deuterio indicano un contributo delle precipitazioni meteoriche nel caso delle falde acquifere sahariane. Hanno anche evidenziato l'effetto di miscelazione delle falde acquifere più profonde dell'acquifero senoniano sottostante. Questo non è il caso delle falde acquifere superficiali delle oasi che sono ricaricate dall'infiltrazione di acque irrigue. L'eccesso di deuterio dimostra che la dissoluzione delle evaporiti è il principale processo di salinizzazione (contribuendo fino al 90%), rispetto all'evaporazione per le falde acquifere dell'oasi. I dati sul trizio indicano la presenza di una ricarica locale nella regione sahariana; il bilancio di massa dei cloruri permette di stimare una ricarica di 13,2 mm/anno. L'uso combinato di carbonio-14 e carbonio-13 mostrano due gruppi principali di acqua: (1) acque degli acquiferi sahariani che mostrano una leggera evoluzione da un termine più antico ad uno recente e (2) acque antiche rappresentate sia dagli acquiferi senoniani che da quelli delle oasi. Un terzo gruppo intermedio evidenzia l'effetto di miscelazione nel caso di numerosi campioni provenienti da acquiferi sahariani adiacenti.
In conclusione, l'indagine basata sugli isotopi degli acquiferi poco profondi può aiutare i decisori a impostare un approccio sostenibile per la gestione delle falde acquifere nell'ambiente ostile di Djerid.

Abstract

Groundwater is a crucial resource for various purposes, especially in the arid land of Southern Tunisia. The assessment of its properties needs a deeper understanding of the main mineralization processes. Therefore, a comprehensive characterization of shallow aquifers in Chott Djerid area has been carried out using isotope methods based on oxygen-18, deuterium, tritium, carbon-14 and carbon-13 on 46 water samples. The obtained results indicated that both oxygen-18 and deuterium pled to meteoric precipitations in the case of the Saharan aquifers. They also highlighted the mixing effect from deeper groundwaters of the underlying Senonian aquifer. This was not the case of the oasis shallow groundwaters which resulted from the irrigation return flow. Deuterium excess showed that the dissolution of evaporites was considered the main process of salinization (contributing up to 90%), compared to evaporation for oasis groundwaters. Tritium data indicated the presence of a local recharge in the Saharan region; it was estimated by the chloride mass balance to be 13.2 mm/year. The combination of carbon-14 and carbon-13 showed two main groups of water: (1) Waters of the Saharan aquifers showing a slight evolution from an old to a recent pole and (2) old waters represented by both the Senonian and the oasis aquifers. A third intermediate group highlighted the mixing effect in the case of several samples from the Saharan neighboring aquifers.
In conclusion, isotope-based investigation of shallow aquifers may help decision makers to set up a sustainable approach for groundwater management in the Djerid harsh environment.

Introduction

In southern Tunisia, groundwater is a major source for supplying everyday needs (Haddaji et al., 2024). This resource is fundamentally important for an arid environment where precipitation is scarce. Thus, a well-thought-out approach is needed for prospective and sustainable use. In this context, numerous researchers have undertaken comprehensive assessments of groundwater from different aquifers, belonging to the Southern part of Tunisia (Dassi, 2011; Gaaloul, 2011; Ayed et al., 2017; Agoubi, 2018; Aouiti et al., 2021; Aydi et al., 2022; Haddaji et al., 2024; Ltifi et al., 2024). For instance, Haddaji et al. (2024) raised concern on groundwater quality assessment and monitoring. Also, Abdelkarim et al. (2024) predicted the effects of climate change on groundwater in arid area of Gabes. They demonstrated that a general decline in precipitation and groundwater recharge is foreseen by 2100s. They recommended immediate action to accurately mitigate groundwater shortage. Similarly, Dhaoui et al. (2022) evaluated groundwater vulnerability to salinization of the southern Menzel Habib area using several indicators. They shed light on the current status and future evolution of groundwater vulnerability to salinization. Thus, without concrete measures, water salinization will become a major problem, further compromising already diminishing water quality. This is applicable for the whole southern districts of Tunisia. Ayed et al. (2017), Ben Brahim et al. (2021), Dassi (2011) have attempted to study groundwater quality by using several techniques like multivariate analysis, hydrogeochemistry and GIS-based tools. Nevertheless, those studies were directed to the deep aquifers of Senonian and Turonian age (i.e., Complex terminal and Continental inetrcalaire). Little concern has been allocated to the Mio-Plio-Quaternary aquifers. Only Tarki et al. (2012) have attempted to highlight the main hydrochemical dynamics of this shallow system. Therefore, special attention was given to this resource in the Djerid area to further highlight the hydrodynamic process that governs the origin, pathway and dominant mineralization processes. These vital water sources sustain life in this region, nourishing oases, supporting agriculture, and serving as a lifeline for human populations. However, the future of these aquifers hangs in a delicate balance. Climate change, increasing water demand, and potential contamination threaten their health and sustainability. These aquifers act as natural reservoirs, storing groundwater that infiltrates from infrequent rainfall events. However, their shallow nature makes them vulnerable to various threats. Increased desertification coupled with high temperatures may further reduce recharge rates. Additionally, human activities such as intensive agriculture and potential wastewater infiltration pose a risk of contamination.

In this context, this geochemical study has been performed to address this pressing need for a deeper understanding of Chott Djerid's shallow aquifers properties. By meticulously analyzing isotopic composition of those groundwater, we can gain valuable insights into its current state and potential vulnerabilities.

Geochemical characterization of water samples is a good approach to reveal clues about the source of the water, the minerals it interacts with along its flow path, and potential contamination sources (Mastrorillo et al., 2018; Ltifi et al., 2024). Stable isotope analysis, as a powerful technique, delves into the variations in the isotopic composition of water molecules. Isotopes of hydrogen (δ^2H) and oxygen ($\delta^{18}O$) act as natural tracers, reflecting the origin of the groundwater (e.g., precipitation, surface water) based on fractionation processes during evaporation and condensation events. This information sheds light on recharge patterns and helps identifying potential sources of contamination. Similarly, isotopes with radioactive decay, such as tritium (^3H) and radiocarbon (^{14}C) offer valuable insights into the age of the groundwater. Tritium, a young isotope, provides information on recent recharge events, while ^{14}C analysis helps estimate the age of older groundwater; this is crucial for understanding flow dynamics within the aquifer system. A comprehensive analysis of the geochemical fingerprint is possible to get key information on the Chott Djerid shallow aquifers. Recharge dynamics can be revealed easily; water origin and the plausible pathways it takes to reach the aquifer. This is crucial for identifying recharge zones and managing water resources effectively. Similarly, high salinity levels pose a significant threat to the usability of groundwater. Geochemical analysis aids in identifying sources of salinity, whether natural (evaporite dissolution) or anthropogenic (wastewater infiltration). This is a valuable information to mitigate salinity issues and ensure long-term aquifer health.

The ratio of ^{14}C to ^{13}C ($^{14}C/^{13}C$) can be used to estimate the age of groundwater, trace its origin, and assess potential contamination sources, as the case in the present study. For instance, higher levels of ^{14}C activity would indicate younger groundwater, while lower levels could indicate older groundwater. Similarly, ^{13}C content can provide clues about the source of carbon, such as from decomposing plants or fossil fuels. By studying both isotopes together, we can easily recognize the history and movement of the studied groundwater.

Materials and methods

Site description

The study area belongs to the northern edge of the Sahara Desert (southwestern Tunisia). It is bordered by the Chott range to the north, the Tebaga mountainous chain to the northeast and the Tozeur uplift to the west; it opens on the "Oriental Erg" southwards (Fig. 1). The presence of a nearby salt depression (i.e., Chott Djerid) pertain the area with an arid to hyper arid climate (less than 100 mm/year precipitation; Dhaouadi et al., 2022). The evaporation rate is about 2500 mm/year in the Saharan zone and 1700 mm/year in the oasis (Zammouri et al., 2007).

Due to this harsh environment, settlements in the region are clustered around the oases bordering Chott Djerid. Most of the remaining lands are barren with only scattered pastoral activities (Kraiem et al., 2012; Kraiem et al., 2014).

Geology and hydrogeology

Previous studies indicated a sedimentary basin showing stratified aquifers with lateral and vertical communications due to lenticular aspects (Dassi, 2011; Sahal and Kamel, 2018).

From geological viewpoint, sedimentary deposition from the Lower to Upper Cretaceous periods can be seen in the outcropping formations of the Tebaga and Dahar Mountain ranges (Fig. 1a). The Mio-Pliocene formations, of continental origin, display a distinctive structure: an intermediate clayey layer intercalated between two sandy units (Besser et al., 2021; Dhaouadi et al., 2021). The uppermost sandy layer, largely outcropping across the region, is the main host of those shallow aquifers. The thickness of the underlying clayey layer reached about 100 meters near Chott Djerid, where it forms the base of the whole system (Mamou, 1979). Those shallow groundwaters were classified into oasis and Saharan aquifers (Kraiem et al., 2014). The latter are supplying drinking water and pasture needs, particularly in the stream (i.e., wadi el hallouf). They differ from oasis aquifers found in clayey sand and gypsum formations (Haj-Amor et al., 2017; Dhaouadi et al., 2022). Both features can be characterized according to their recharge modes and piezometry (Fig. 1b). Shallow aquifers of the studied area exhibit subsurface flow from south and east towards Chott Djerid. The Dahar chain, with its significant piezometric gradient, plays a crucial role in recharging these aquifers, particularly near the piedmont. Moreover, the Tebaga chain contributes to recharge, though to a lesser extent. Chott Djerid and the southwestern Douz area serve as the primary discharge areas (Kraiem et al. 2014). The oasis shallow aquifers are supplied with the return flow (i.e., irrigation waters) pumped from the underlying complex Terminal aquifer in the Nefzaoua (The southern part of the map, which includes the region located between Chott Djerid and the Dahar range) and the Djerid regions (covering the geographical area that lies between the western edge of Chott Djerid and the Tozeur Ridge; Dassi, 2011; Tarki et al., 2012).

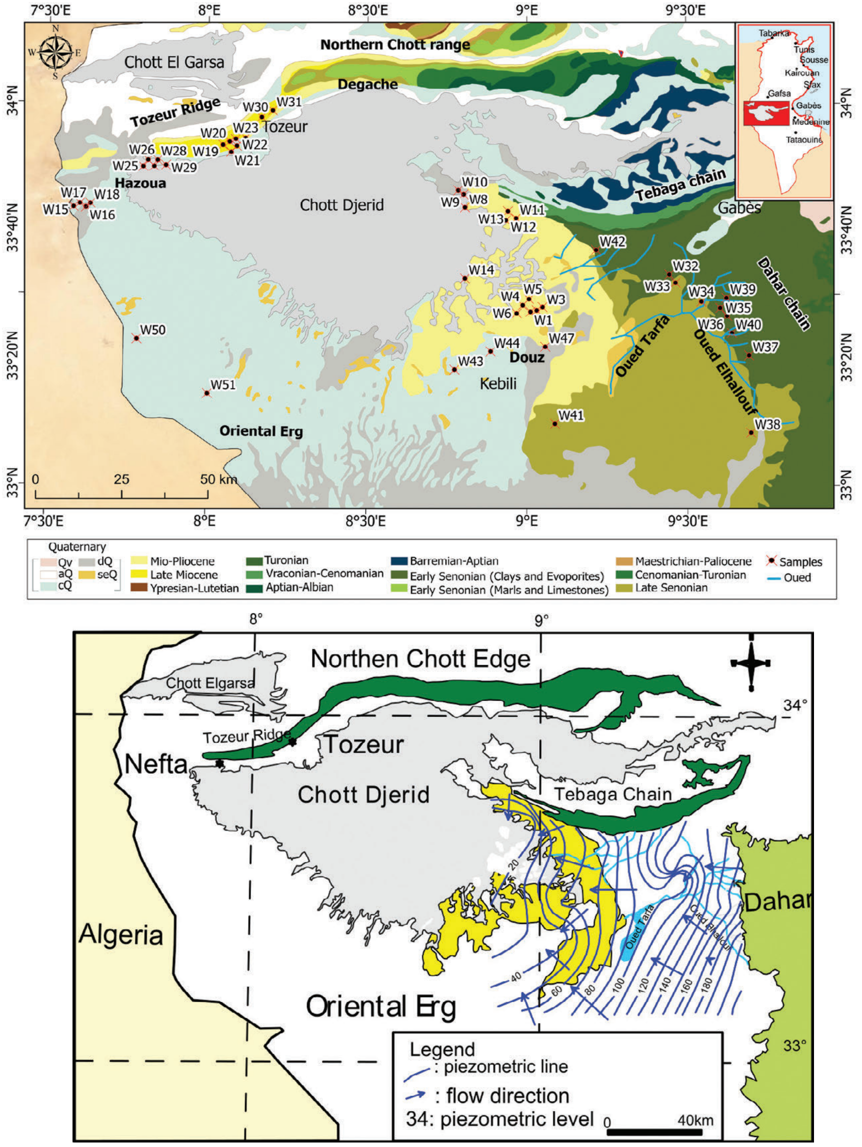

Fig. 1 - Location (a) and piezometric (b) maps of the studied area showing the sampling points.

Fig. 1 - Carta geologica (a) e piezometrica (b) dell'area di studio con indicazione dei punti di campionamento.

Sampling and analysis

Water samples from the Nefzaoua and Djerid shallow aquifers were analyzed for both stable (oxygen 18, deuterium and carbon 13) and radioactive isotopes (Tritium, carbon 14; Hamed et al., 2014). In addition, total dissolved solids (TDS g/L), was measured at the laboratory by a gravimetric method through weighing the residual solids after evaporation of a given amount of the studied water samples. We used a RADWAG type ASR2 Plus precision balance (RADWAG Balances and Scales, La Balance, Tunisia).

Those analyses interested both oasis and Saharan shallow aquifers. Prior to sampling, each water well was purged for 30 minutes. Water samples were collected and filtered through 0.45-micron filters to remove particulate matter (Liu et al., 2023). The filtered samples were stored in pre-cleaned glass vials and immediately sealed to prevent isotopic fractionation due to evaporation. We measured the stable isotopes oxygen-18 (^{18}O) and deuterium (^{2}H) in water samples using laser spectrometry. The results are reported in the standard delta (δ) notation relative to Vienna Standard Mean Ocean Water (VSMOW; Liu et al., 2023). This notation calculates the difference between the isotopic ratio of the sample and the VSMOW standard, multiplied by 1000. The typical precision of our measurements is ±0.1% for ^{18}O and ±1.0% for ^{2}H (Abid et al., 2010). Those stable isotopes remain unchanged as the water moves through the aquifer. Thus, they are not significantly altered by hydrochemical processes within the aquifer. Therefore, their signature can reveal the source of the water.

Tritium analyses were performed using electrolytic enrichment and liquid scintillation spectrometry (Morgenstern and Taylor, 2009). Tritium concentration was expressed in Tritium Units (TU), where 1 TU is defined as the isotope ratio ^{3}H/^{1}H = 10-18.

In-situ operation consisted in precipitating the dissolved carbon by hydrated barium chloride ($BaCl_2$, $2H_2O$) after adjusting pH to the desired value (i.e., 10-11) with 1M NaOH. The sample was then immediately stored to avoid CO_2 absorption. In addition, the natural abundance of carbon in groundwater consists of two main isotopes (i.e., ^{13}C and ^{14}C). ^{13}C is a stable isotope while ^{14}C undergoes radioactive decay with a half-life of approximately 5,730 years. Analysis of ^{13}C and ^{14}C content of groundwater can provide valuable insights into various environmental processes. The activity of carbon 14 is measured by scintillation counting on benzene (Molnár et al., 2022; Stojkovic et al., 2024).

^{14}C activities are expressed as a percentage relative to modern carbon (pcm). They correspond to the apparent ages or residence times, expressed in years Before present (B.P.). Values of δ^{13}C were expressed versus the Vienna Pee Dee Belemnite (VPDB) standard. The precision of measurements for stable isotope and radioactive analysis was ±0,1‰ for δ^{18}O, ±1‰ for δ^{2}H, ±0.3 TU for ^{3}H and ±0.3‰ for ^{13}C.

Results and discussion

Isotope study can be a powerful tool for deciphering the southern shallow aquifers properties (Table 1; Table 2). All the samples are groundwaters. Irrigation waters come from the confined underlying Senonian aquifer (CT). That is why mixing with shallow groundwater may change the properties of the samples studied. Analyzing the ratios of stable isotopes like oxygen-18 and deuterium in groundwater samples is a crucial step for understanding groundwater flow patterns and the whole aquifer system. In addition, one can easily recognize past rainfall patterns and drought cycles to retrace the past climate conditions in the Djerid area.

Salinity demonstrated a heterogeneous distribution, as shown in Figure 2. Similarly, TDS values exhibit low values (around 2.5 g/L) for water samples from Oued el Hallouf and specific locations in southern Nefzaoua (i.e., W38, W37, W40, W36, W39, W35, W34, W41, W44, W43). Groundwater samples with lower salinity, typically below 2 g/L, correspond to the recharge zone, as indicated by the piezometric map (Fig. 2). Conversely, the remaining samples exhibit higher salinity, ranging from 2 g/L to 11 g/L. These high-salinity waters are mainly found in shallow Saharan aquifers and oasis aquifers. The observed heterogeneity in salinity values may be attributed to various factors, including the dissolution of evaporites (halite, gypsum, anhydrite, and sylvite), the accumulation and infiltration of saline water resulting from excess irrigation, and the effects of evaporation. Salinity may also be attributed to the intrusion of drainage water from neighboring areas or basins near oasis shorelines (Kraiem et al., 2012).

Stable isotopes of oxygen 18 and deuterium

Both groundwater and irrigation water samples from the Nefzaoua area were represented on the diagram δ^{2}H/δ^{18}O; they showed three mains groups (Fig. 3a): (1) A first group (Group 1) included water samples above the global and local meteoric water lines (GMWL and LMWL, respectively) with an average isotopic composition equal to that of rainwater in the Nefzaoua region (i.e., -5 ‰ VSMOW for ^{18}O and -30‰ VSMOW for ^{2}H). These water samples are from recent recharge via direct infiltration of rainwater. This group mainly characterizes groundwaters of Wadi El Hallouf, at the foot of Dahar, where recharge can take place (Fig. 1b); (2) A second group (Group 2) composed of samples representing waters from the oasis shallow aquifers with a very close similarity to those of irrigation water, probably due to the return of irrigation water and (3) A third group (Group 3) gathered representative water samples from the Saharan aquifers located towards the South of chott Djerid. A plausible explanation of those water properties is the effects of mixing between different types of water. As expected, only a few points, located in the evaporation area, are indicative of high evaporation.

Tab. 1 - Isotopic characteristics of the studied water samples.

Tab. 1 - Caratteristiche isotopiche dei campioni d'acqua studiati.

Well N°	^{18}O (‰VSMOW)	^{2}H (‰VSMOW)	^{3}H (TU)	Activity^{14}C (%)	δ^{13}C (PDB)	TDS (g/L)
W1	-6.08	-47.84	0.18	60±2%	-12.46	10.69
W2	-3.78	-34.28				9.97
W3	-5.74	-46.96	0.61			6.55
W4	-5.58	-46.29				9.10
W5	-1.86	-25.89	0	97±1.8%	-7.41	9.29
W6	-4.86	-41.98	0.31			8.05
W8	-4.37	-43.25	0	80±1.7	-12.46	7.13
W9	-7.10	-54.19		105.4±1.2	-8.46	6.03
W10			0			4.96
W11			0			6.23
W12	-5.89	-48.20		108.8±1.5	-14.24	5.99
W13	-5.95	-49.13				7.54
W14	-5.62	-44.12	0.14	60.4±1.8	-12.37	4.98
W15	-1.58	-29.71	0.5			6.99
W16			0.4			7.12
W17	-2.67	-36.05				6.80
W18	-5.20	-42.73	0.3			7.77
W19	-4.10	-43.59	0			6.68
W20	-4.14	-43.24				6.76
W21	-4.88	-37.41	0.6			9.26
W22	-4.47	-41.35	0.5			7.34
W23	-3.34	-34.35	0.3			9.07
W24			0.2			8.14
W25			0			8.17
W26	-5.06	-45.10	0.5			6.12
W27	-3.55	-42.08	0.6			6.26
W28	-5.04	-43.72	0.7			6.88
W29	-5.02	-44.44	0.2			6.14
W30	-5.83	-42.33	0			7.60
W31	-4.96	-40.75	0.2			11.01
W32	-4.41	-32.21	0.37			7.85
W33	-5.47	-33.80	0.38	20.7±3	-5.57	3.70
W34	-5.70	-34.38				0.92
W35	-5.75	-33.76	3.51			0.56
W36	-5.55	-31.90		99.5±1.2	-8.61	0.36
W37	-5.61	-31.70	3.23	78.3±2%	-5.01	0.40
W38	-6.47	-40.40				1.86
W39			3.85			2.56
W40	-4.81	-25.80				0.51
W41	-4.76	-35.07				0.45
W42	-5.16	-38.44	1.50			4.11
W43	-5.16	-42.90		52±1.5	-5.89	2.26
W44	-5.39	-44.43				1.64
W47	-4.97	-41.90	0.00			4.73
W50				107±1.7	-10.51	7.22
W51				87± 1.8	-7.53	4.61

Tab. 2 - Summary statistics of the main isotopes.

Tab. 2 - Statistici riassuntivi dei principali isotopi.

Variables	mean	sd	median	mad	min	max	range	skew	kurtosis	se
Depth (m)	13.71	13.22	8	2.97	3.9	50	46.1	1.84	2.07	2.06
^{18}O (‰) VSMOW	-4.89	1.15	-5.05	0.91	-7.1	-1.58	5.53	1.05	1.15	0.19
^{2}H (‰) VSMOW	-39.89	6.56	-41.94	6.58	-54.19	-25.8	28.39	0.29	-0.55	1.06
^{3}H (TU)	0.94	1.4	0.31	0.46	0	3.85	3.85	1.17	-0.42	0.36
Activity ^{14}C (%)	79.67	26.98	83.5	33.36	20.7	108.8	88.1	-0.7	-0.65	7.79

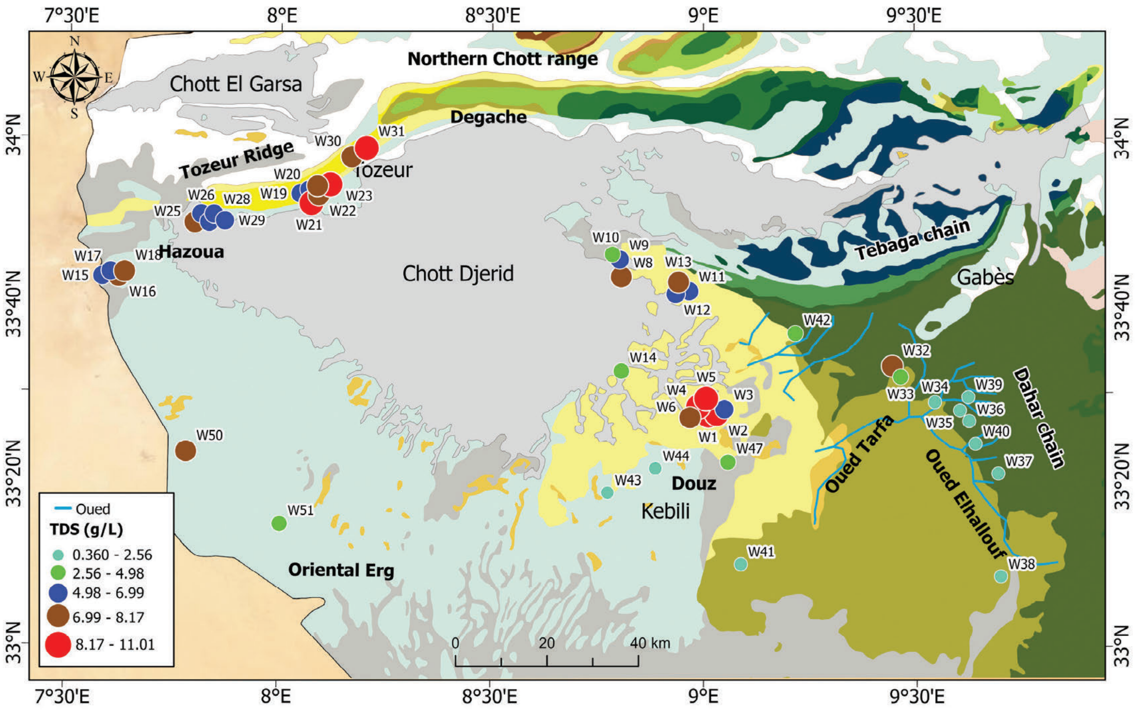

Fig. 2 - Salinity map distribution of water samples in the study area.

Fig. 2 - Carta della distribuzione della salinità dei campioni d'acqua nell'area di studio.

For the Djerid area (Fig. 3b), the correlation diagram of deuterium contents versus oxygen-18 showed that both oasis and irrigation waters are below the GMWL and LMWL. Samples collected in the oasis aquifer form a homogeneous group with average isotopic composition comparable to that of irrigation water. It was also noticed that some points clustered along the evaporation line of slope 3, corresponding to the evaporation that probably took place during the infiltration of irrigation water. There is a clear tendency for hydrogen and oxygen isotopes to appear on the right side of the biplot. This can be attributed to the effect of evaporation, which preferentially removes lighter isotopes (deuterium and oxygen-18) from water bodies, leaving behind a residue enriched in heavier isotopes. This enrichment can shift the isotopic composition of the water towards the right side of the biplot. Additionally, other processes such as water-rock interaction and mixing with different water sources can also contribute to the observed isotopic patterns.

Spatial distribution of $\delta^{2}H$ and $\delta^{18}O$ isotopes suggested a northwest-to-southeast flow pattern in the Nefzaoua region, originating in the mountainous areas and moving towards Chott Djerid. This pattern can be attributed to processes such as evaporation or mineral-water interactions. It was also revealed that groundwater in Wadi El Hallouf and southern Nefzaoua exhibited higher concentrations of oxygen-18 and deuterium. Conversely, shallow aquifers of the oases display lower concentrations of these isotopes (Fig. 4).

Importance of deuterium excess

Understanding the origin and mineralization of groundwater in particular environments is crucial in determining the prevailing hydro-geochemical process. Deuterium excess, a measure of the enrichment of heavier deuterium isotope in water compared to seawater, acts as a fingerprint to retrace the water's source (e.g., precipitation, evaporation) and identify the mechanisms that led to its mineralization (Hassen et al., 2016). For instance, high deuterium excess indicate low evaporation, suggesting that the water originated directly from precipitation (Hepp et al., 2015). Conversely, low values are indicative of significant evaporation, potentially concentrating minerals in the remaining water.

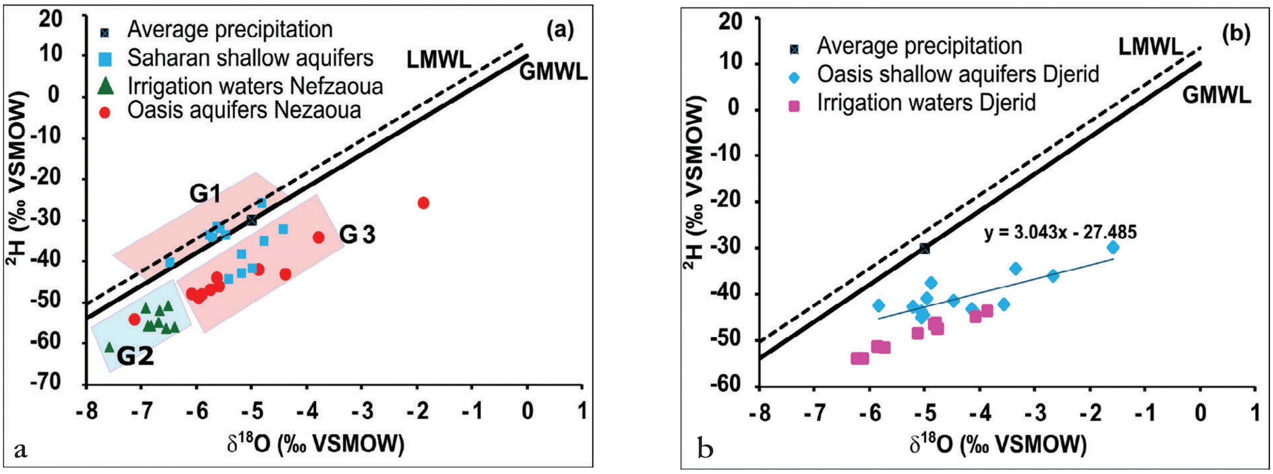

Fig. 3 - δ^2H versus δ^{18}O of groundwater in the Nefzaoua (a) and Oasis of Djerid (b).

Fig. 3 - Grafico δ^2H versus δ^{18}O delle acque sotterranee a Nefzaoua (a) e Oasi di Djerid (b).

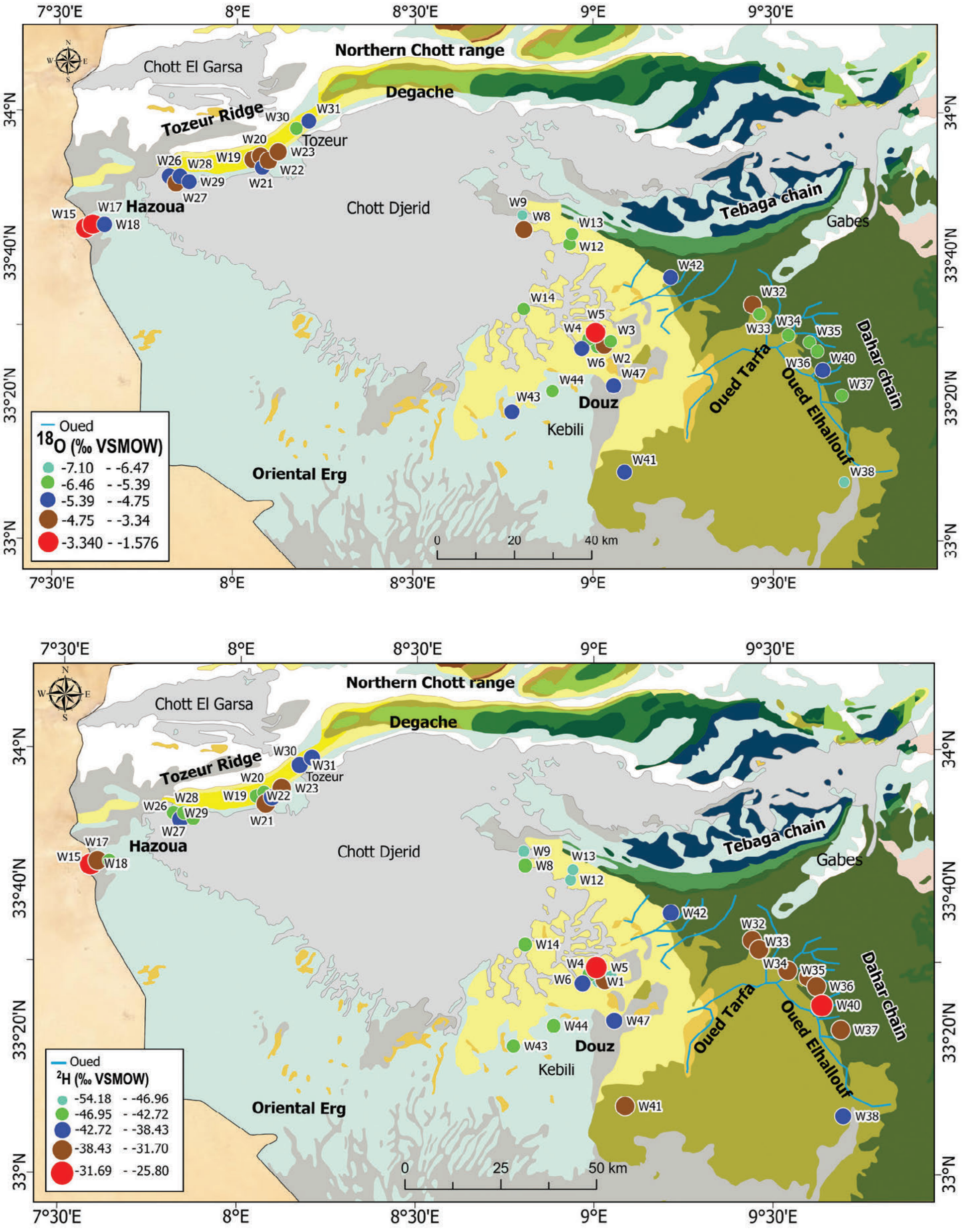

Fig. 4 - Distribution map of Oxygen-18 (a) and deuterium (b) contents of the studied groundwater samples.

Fig. 4 - Carta della distribuzione di Ossigeno-18 (a) e Deuterio (b) nei campioni di acque sotterranee studiati.

Stable isotope composition can be determined from the equation (1):

$$\delta^2H= 8\delta^{18}O+10 \qquad (1)$$

When a precipitation water is subjected to evaporation during the recharge of rivers, lakes and underground aquifer by infiltration from the soil, the slope of the evaporation line will be less than 8 (i.e., 4 - 6 for open water and 2 -3 for soil evaporation; Ren et al. 2024). There should be no isotope fractionation during drainage, plant transpiration (Xu et al., 2020) and dissolution of minerals, probably because vegetation excludes salt during water absorption, transpiration and mineral dissolution. Therefore, increasing the salinity of residual water would take place without isotope fractionation. Similarly, when a water table level is deep and stable, groundwater should evaporate persistently from capillary moisture without causing isotope fractionation in the remaining groundwater. Nevertheless, an obvious isotopic fractionation may take place if the water body is subjected to a high evaporation rate, especially in lakes, water reservoirs and sebkhas.

Therefore, the relationship between groundwater salinity and $\delta^{18}O$ (or δ^2H) cannot be used to demonstrate the evaporation process, since the isotopic compositions of the initial water (precipitation or mountain rivers) is usually influenced by seasonal variations. To overcome such a shortcoming, Dansgard (1964) proposed the use of d-value to describe the excess deuterium of global precipitation. The d-value is defined for a slope of 8 and calculated for each water sample as follows:

$$d= \delta^2H-8\delta^{18}O+10 \qquad (2)$$

d is estimated to 10‰ on average for precipitation. When a water body undergoes evaporation, the excess deuterium will decrease, and the salinity will increase. By combining the notation δ and Rayleigh distillation, the relationship between the excess of deuterium (d) and the remaining fraction of the aquifer (f) can be obtained by determining the excess of deuterium (d) (Eq. (6)).

$$\delta=(R/R_{VSMOW}-1)\ *1000 \qquad (3)$$

$$R=R_0f\ ({}^{\alpha}{}_{v-1}{}^{-1}) \qquad (4)$$

$$\delta= ((R_{0f}{}^{(\alpha-1)}/R_{VSMOW})-1)\ *1000=[(\delta_0/1000)+1)\ f^{(\alpha-1)}-1]\ *1000= (\delta_0+1000)\ f^{(\alpha-1)}-1000 \qquad (5)$$

$$d=\delta^{2H-8\delta^{18}O}=(\delta_0{}^2H+1000)f^{(\alpha 2H-1)}-8(\delta_0{}^{18}O+1000)f^{(\alpha^{18}O-1)}\ 7000 \qquad (6)$$

where R is the isotope ratio; f is the remaining fraction of the aquifer; $\alpha(\alpha_{v-l})$ is the fractionation factor between vapor and liquid water; $\delta^{18}O$ and δ^2H are the δ values of the initial water. The total isotope fractionation factor α_{l-v} $(1/v_{-l})$ of free water and air is equal to the sum of the equilibrium fractionation factor for vapor-water exchange (ε_{l-v}) and the kinetic factor ($\Delta\ \varepsilon_{Bl-v}$) (Clark et Fritz, 1997).

With given evaporation conditions (i.e., temperature and relative humidity), the ratio between d and f can be obtained by Eq. (6). Here, the $\delta_0{}^{18}O$ and $\delta_0{}^2H$, unknown in the Eq. (6), are of no concern until the values of the deuterium excess are set for the initial water (on average 10‰).

To determine the dissolution (%), evaporation and the remaining fraction of the aquifer affecting the sampled waters in the study area, the initial water parameters must be defined and fixed.

Definition of initial water parameters

In the study area, the initial water is represented by irrigation water or water from the Complex Terminal (CT). The complex Terminal aquifer is a multilayered reservoir. For this study, we mentioned only the Senonian formation for the Nefzaoua region and the Miocene formation for the Djerid region as irrigation waters. This water itself comes from rainwater. As it passed through the aquifer formations, it acquired its salinity by dissolution of the encompassing layers. This water is known for its oxygen-18 and deuterium isotopic composition of about -5‰ VSMOW and-30‰ VSMOW, respectively, a deuterium excess (d0) of 10‰ vs. SMOW and an initial salinity S0 of 0.13 g/L.

According to carbon 14 correspondence (%) and relevant literature of the Saharan platform and North Africa (Edmunds et al., 1997; Guendouz et al., 1997), Holocene was proposed as an ultimate period of groundwater infiltration from the CT aquifer (Kamel et al., 2008). This period was characterized by a warming of 3°C associated with an enrichment of 3‰ in $\delta^{18}O$ (Annalisa, 2007). Such enrichment was associated with a significant variation in deuterium excess that varied between ±0.3‰ for a variation of 0.5°C for the sampling site and 1°C for the initial water temperatures (Masson-Delmotte et al., 2005). A sensitivity study on the variation of deuterium excess versus Holocene precipitation showed that, for a warming of 3°C, an increase in deuterium excess of about 1‰ can be observed. These variations were independent of oxygen-18 and deuterium isotopic composition of the original water.

Determination of dissolution and evaporation

Huang & Pang (2012) proposed an evapo-concentration model for drylands. This model is based on the fact that the salinity of water doubles when half of the water is evaporated (Huang and Pang, 2012). With given temperature and humidity conditions (Nefzaoua: T = 21.4°C and H=58% and Djerid: T = 21.8 and H= 51%, a salinity S0=0.13 g/L and a d_0=10‰VSMOW; Kraiem 2015), it is possible to calculate the deuterium excess (d), based on equation (6). The evolution of d as a function of f and S is illustrated in Figs. 5a and 5b, for Nefzaoua and Djerid, respectively.

The correlation of d versus S allowed the determination of the characteristic evapo-concentration and dissolution domains for each region (Fig. 6; Huang & Pang, 2012). For a given water body with an initial salinity, the remaining fraction after evaporation (f) should have higher salinity as evaporation directly contributes to the salinity. Mineral dissolution and/or transpiration also contributes to salinity S (which is represented by the total dissolved solids or TDS (g/L),

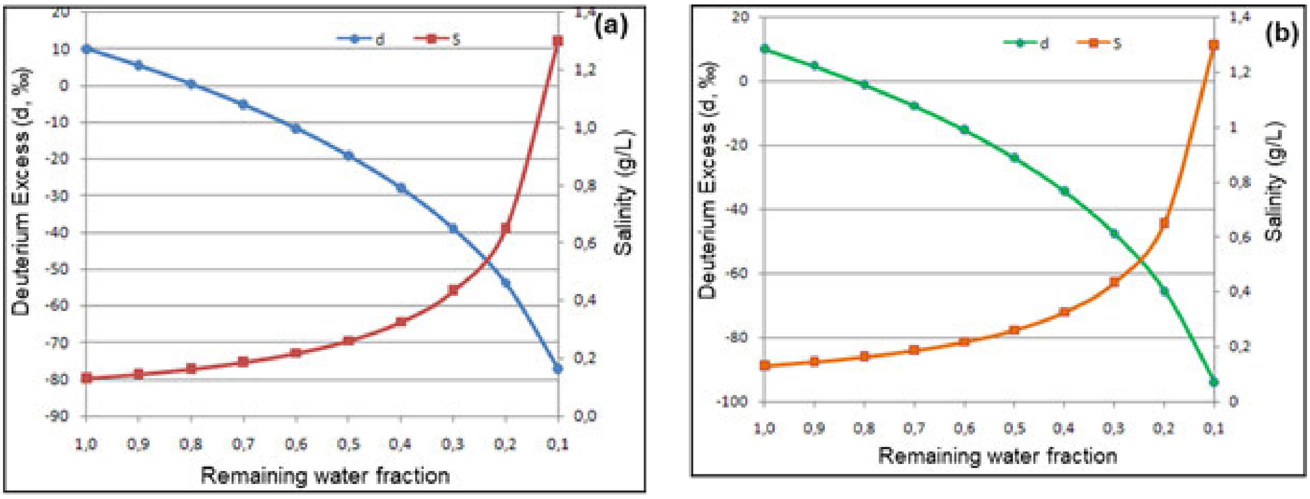

Fig. 5 - Variation of deuterium (d) and salinity (S) versus the fraction of the remaining water (f) during evaporation with an initial d excess (d) of 10‰, an initial salinity of 0.13 g/L, a temperature of 21.8°C (a-Nefzaoua, humidity 58%; b Djerid, humidity 51%)

Fig. 5 - Variazione del Deuterio (d) e della salinità (S) rispetto alla frazione di acqua rimanente (f) durante l'evaporazione con un eccesso iniziale di Deuterio del 10‰, una salinità iniziale di 0,13 g/L, una temperatura di 21.8°C (a-Nefzaoua, umidità 58%; b Djerid, umidità 51%).

(Table 1). The contribution of evapo-concentration to salinity and the contribution of mineral dissolution and/or transpiration to total salinity can be calculated based on Huang & Pang (2012).

Figure 6 shows the correlations between salinity and deuterium excess for drainage water and groundwater sampling points, respectively, at the Nefzaoua and Djerid areas. From the slope of the characteristic evapo-concentration line for each region, the rate of evaporation E and dissolution (d) and the fraction f for each water point can be determined. Dissolution contributes with a significant percentage (from 90 to 98%) and that evaporation, even if it exists, remains minimal compared to the dissolution that took place during the infiltration of water into the aquifer in contact with the host rock.

In fact, the calculations made for the rate of dissolution and evaporation with an excess of deuterium of the initial water of 10 ‰VSMOW show that the dissolution participates with 90% compared to the evaporation which remains minimal. This shows that, even considering the variations of the deuterium excess versus temperature, it can be seen that the margins of variation of the dissolution and evaporation rates vary with only 3 to 4%, indicating that dissolution is the main process of salt charge acquisition.

Therefore, despite the aridity of the climate and the importance of evaporation as a mineralization process at the oasis aquifers. Thus, dissolution should be the main process of the acquisition of higher salt content, participating with a rate of 90%.

Recharge evaluation based on tritium isotope and chloride content

Tritium, a radioactive isotope of hydrogen, acts as a tracer in water systems. By analyzing its concentration (measured in Tritium Units or TU) across various locations, valuable

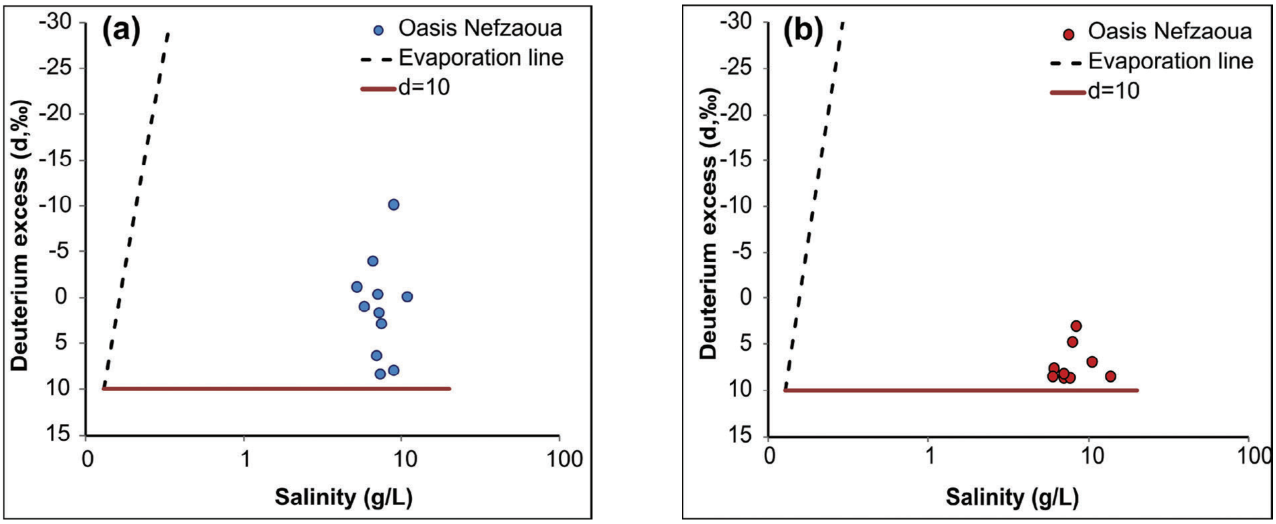

Fig. 6 - Correlation diagram of the deuterium excess versus salinity of water samples from the oasis aquifers in the Nefzaoua (a) and Djerid (b).

Fig. 6 - Diagramma di correlazione tra l'eccesso di deuterio e la salinità nei campioni d'acqua provenienti dagli acquiferi delle oasi di Nefzaoua (a) e Djerid (b).

insights into the age of the water, water source and mixing and groundwater flow patterns among others (e.g., anthropogenic effects and climate change) can be proposed. Tritium is a young isotope, with a half-life of 12.3 years. Higher tritium levels indicate the presence of younger water, potentially reflecting recent infiltration from rainfall or surface water sources. Conversely, lower tritium content suggests older groundwater that has been distinguished from recent recharge events (Pourcelot et al., 2013).

Variation in tritium content also indicate the contribution of different water sources to a particular location, as is the case of the south Nefzaoua area. For instance, groundwater with higher tritium might be mixed with recent rainfall input.

The main direction and speed of groundwater flow can be inferred from the spatial distribution of tritium. Areas with decreasing tritium content along the flow path suggest progressive dilution with older, tritium-depleted water.

It was stated that Past nuclear activities released fraction of tritium as an imprint on the spatial distribution (Pourcelot et al., 2013). Elevated tritium levels in unexpected locations might point towards past contamination events.

By combining spatial tritium data with other environmental isotope techniques and hydrogeological information, a comprehensive picture of the water dynamics within a system can be built. This is fundamental for sustainable water resource management, pollution source identification and climate change impact assessment to get insights into how climate variations affect groundwater recharge patterns (Gourcy et al., 2022).

The spatial distribution map of tritium contents showed significant spatial variations from the eastern to western edges of the study area and from the piedmonts of Dahar towards the Chott (Fig. 7). Tritium contents of 3 and 4TU characterized the water samples of the Wadi El Hallouf and Dahar. Similarly, values of 1TU and 2TU were seen in the Tebaga area, similar to those of current rainfall, indicating a significant contribution of rainwater in groundwater recharge. Far away from the wadis, a clear decrease in the tritium contents to reach ±0.8TU can be attributed to an extremely low recharge from recent precipitation waters in the context of higher relief sceneries (Rapti-Caputo and Martinelli, 2009).

Figure 8 coincides with the ^{3}H distribution map and makes it possible to classify the waters into separate groups:

- Waters tritium-free with contents <1TU and depleted in stable isotopes (G1). It is the ancient, paleo-climatic waters taken from the CT aquifer and those from oasis aquifers that constitute the return of irrigation of water from this aquifer. This group represents most of the sampled points. It should also be noted that this group includes the representative points of wells which constitute part of the Saharan groundwater. These are points that seem to receive a strong contribution from ancient waters from the CT aquifer masking any recharge effect in these areas.
- A group of waters with contents <1TU and enriched with stable isotopes (G2). This group represents only few points. These are waters from the return of irrigation of deep aquifers, but which have had evaporation during

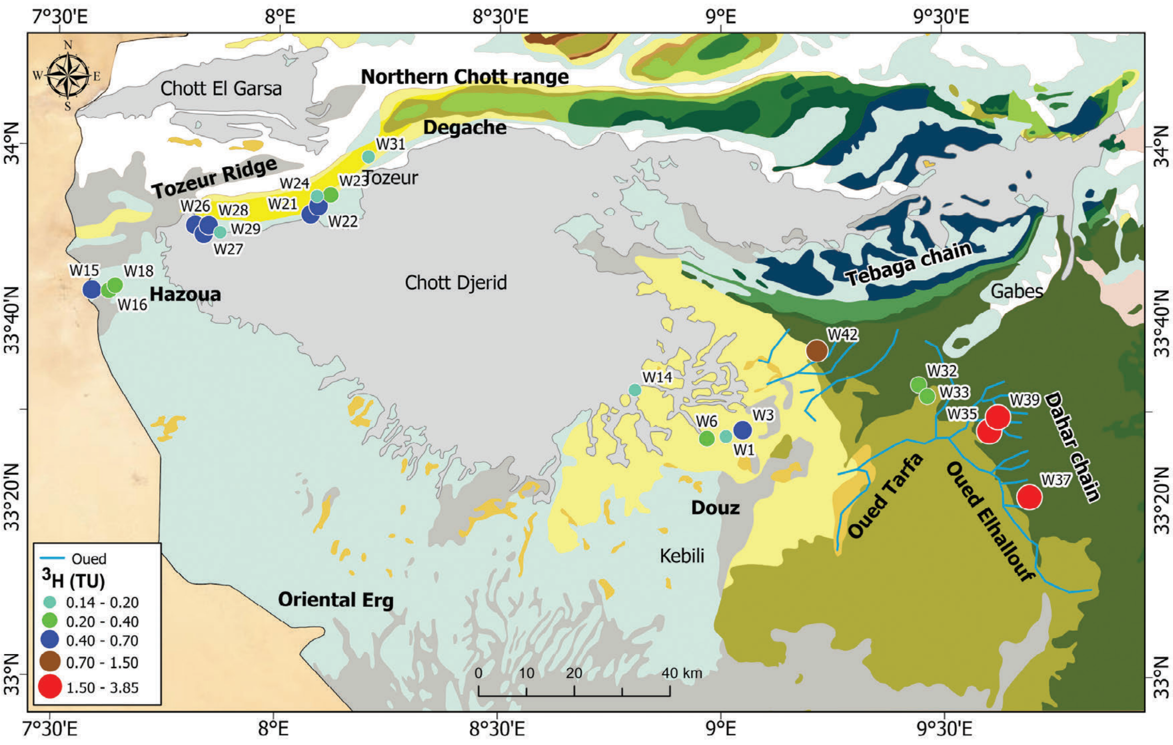

Fig. 7 - Spatial distribution of tritium contents in the studied area

Fig. 7 - Distribuzione spaziale del contenuto di Trizio nell'area studiata.

their infiltration at the level of the aquifers. These low tritium levels could account for two phenomena: a low or even the absence of a current recharge by rainwater at these localities and the existence of a significant contribution of water from the return flow of old origin thus masking any possible recent recharge.

- A group of waters enriched in tritium with contents between 1 TU and 4TU and with variable levels of stable isotopes (G3). These are the recent waters taken from the surface wells of the Plio-Quaternary aquifer. These waters indicate tritium levels similar to those of precipitation water at the Sfax weather station between 1992 and 1998 and probably correspond to charging during the last decade. In fact, the highly variable levels of stable isotopes in these waters indicate the interference of various phenomena such as the mixing process with ancient waters with depleted stable isotopes contents and the evaporation process, characterizing a relatively slow space recharge.

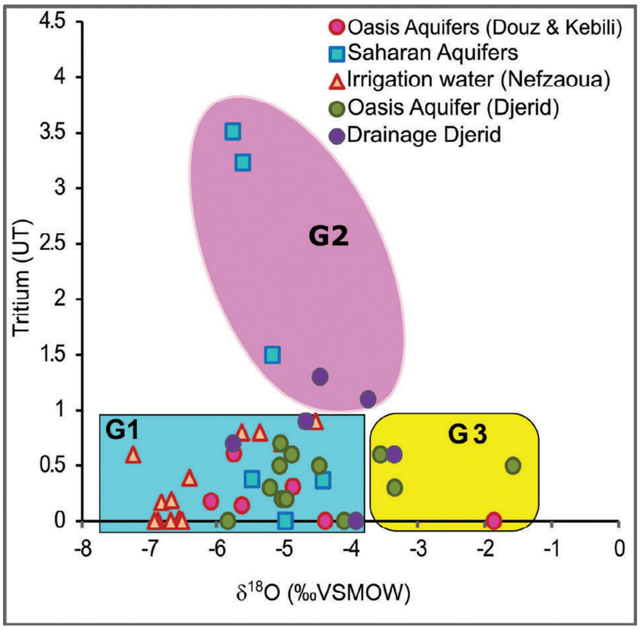

Fig. 8 - Tritium versus oxygen-18 concentrations in the groundwater samples studied.

Fig. 8 - Concentrazioni di trizio e ossigeno-18 nei campioni di acqua sotterranea studiati.

Chloride balance

The chloride ion is a highly soluble, non-absorbent, chemically conservative, and easily measurable environmental tracer. It has been used to estimate recharge in arid and semi-arid areas for more than two decades (Wilske et al. 2020; Boumaiza et al., 2021; Edmunds & Tyler, 2002). According to Liu et al. (2024), in several types of rocks, a significant correlation exists between precipitation and chloride content, suggesting rainfall as a recharge function. In addition, most plant species do not take significant amounts of chloride from soil water while participating in their concentration through evapotranspiration in the root zone (Boumaiza et al., 2021). As a result, the chloride content in groundwater is usually proportional to the recharge as indicated by Liu et al. (2024):

The chloride balance was applied to estimate recharge in the current study area. The weighted average concentration of chloride in precipitation was estimated to be 51.6 mm.

In fact, the recharge mechanism differs from the oasis to the Saharan environment. Concerning oasis aquifers, chloride has different origins, since recharge is ensured, largely by irrigation water with the possibility of returning and intrusion of drainage water to oasis aquifers (Kraiem et al., 2012), and to a lesser extent by rainwater in exceptionally rainy periods. This limits the possible use of chloride balance in the determination of recharge in the oasis aquifers. Recharge of Saharan aquifers is primarily driven by rainwater infiltration. It can be quantified using the following formula:

$$R=(P*Cl\ rain)/cl\ well \qquad (7)$$

With R: the average annual recharge (mm); Cl aquifer: the average concentration of chlorides in the aquifer (mg/L); P: the average annual precipitation in the study area in (mm); Cl rainfall: the weighted average content of chloride concentrations in precipitation (mg/L).

The calculation results show R values varying between 68 mm and 0.5 mm with an average of 13.2 mm (Table 3). This is clearly demonstrated by the fact that recharge is high at wells with high tritium activity (i.e., W35, W37 and W39; Fig. 1) and less important at water points with the lowest tritium activities (e.g. wells 32 and 47).

Tab. 3- Chloride contents (mg/L), recharge and tritium (TU) content of Saharan aquifer wells.

Tab. 3 - Contenuto di cloruro (mg/L), ricarica e contenuto di trizio (TU) dei pozzi dell'acquifero sahariano.

Well number	Cl (mg/L)	R (mm/y)	3H (TU)
W32	1562.3	1.1	0.4
W 33	354.1	5.1	0.4
W 34	94.9	18.9	-
W 35	36.1	49.6	3.5
W 36	26.4	68.0	-
W 37	34.8	51.5	3.2
W 38	289.2	6.2	-
W 39	95.3	18.8	3.9
W 40	31.0	57.8	-
W 41	104.3	17.2	-
W 42	624.8	2.9	1.5
W 43	601.3	3.0	
W 44	301.8	5.9	
W 45	1207.0	1.5	
W 46	3337.0	0.5	
W 47	1518.0	1.2	0.0
W 48	356.7	5.0	
W 49	1741.3	1.0	
W 50	2512.4	0.7	
W 51	1072.4	1.7	
W 52	983.4	1.8	

Carbone 14 and Carbone 13 isotopes

Radiocarbon (^{14}C) dating is a scientific technique used to determine the age of organic matter by measuring the amount of ^{14}C. This technique is particularly useful for dating groundwater in shallow aquifers.

Groundwater in shallow aquifers is usually juvenile with higher ^{14}C activity due to its shorter residence time. This means that it has been in contact with the atmosphere more recently where it can absorb ^{14}C produced by cosmic rays. As the depth of the aquifer increases, the residence time of the groundwater also increases. This means the groundwater has been isolated from the atmosphere for a longer period and has had more time for its ^{14}C to decay. Consequently, deeper groundwater will have lower ^{14}C activity. Thus, measuring the ^{14}C activity of groundwater samples from different depths within a shallow aquifer become possible to estimate the age of the groundwater and assess how long it has been since its recharge by infiltration. This information is valuable for understanding groundwater recharge rates, flow patterns, and potential contamination risks.

Nevertheless, ^{14}C activity in groundwater can be directly affected by several parameters, including the host rock lithology, biological and anthropogenic activities. Local geology and soil through which the groundwater flows strongly affect ^{14}C activity. Similarly, microorganisms in the aquifer can consume ^{14}C, lowering its activity in the groundwater. Moreover, anthropogenic activities have increased the amount of ^{14}C in the atmosphere in recent decades. This can make shallow groundwater appear younger than its real age.

By considering all these factors, scientists can use ^{14}C dating to obtain a more accurate understanding of the age and origin of groundwater in shallow aquifers. Within the study area, twelve samples were collected for carbon-14 analysis. These carbon-14 activities were subsequently plotted on an activity map, revealing a heterogeneous distribution across the entire region (Fig. 9). Most of surface sinks showed variable carbon-14 activities, ranging between 80 and 100%. This is a result of freshwater precipitation, with exceptions for sample W43 and W33 (average activities were 52 and 20.7%, respectively). Such a deviation from ordinary values measured at the surface wells can be attributed to the mixing effect of older/deeper waters. This corroborates well with findings of Hamed et al. (2014) who investigated the potential sources of groundwater contamination and the hydrodynamic functioning of the multilayer aquifer system in the Gafsa area (Southern Tunisia). In fact, shallow waters may have been stored during rainy periods of the Plio-Quaternary recognized as the main groundwater recharge period in southern Tunisia (Guendouz et al., 1997; Edmunds et al., 2003).

Oxygen-18 contents versus carbon-14 activity showed that surface wells stored fresh water with extremely high ^{14}C activities, especially the water samples from the Nefzaoua area (Fig. 10). These waters are recent generated from the infiltration of precipitation water, as in the case of Saharan aquifers. Another plausible explanation resides in the return of irrigation water after carbon exchanges with atmospheric CO_2. This will contribute to the enrichment of heavy carbon concomitantly with a depletion in light carbon (the case of oasis aquifers). We distinguish, in fact, irrigation water forming a group of old waters. However, the waters of oasis aquifers indicated a rejuvenating effect associated with exchanges with CO_2 from the soil with an ancient origin materialized by irrigation water. These have been dated Late Pleistocene/Holocene (Guendouz et al., 1997; Abid et al., 2010). The waters of saharan aquifers showed a slight evolution from an old to a recent pole.

These representative points of groundwater were plotted on a diagram representing the variation of carbon-14 activities versus depth (Fig. 11). The activity decreased with depth, highlighting the effect of exchanges with soil CO_2 for the

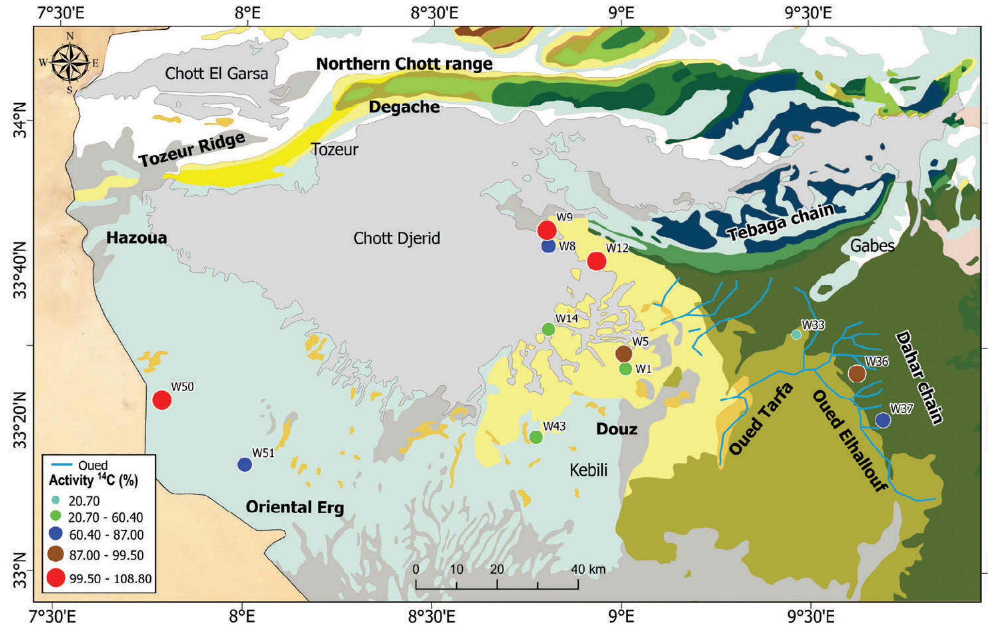

Fig. 9 - Spatial distribution of carbon-14 activities of groundwater samples

Fig. 9 - Distribuzione spaziale dell'attività del carbonio-14 nei campioni di acque sotterranee.

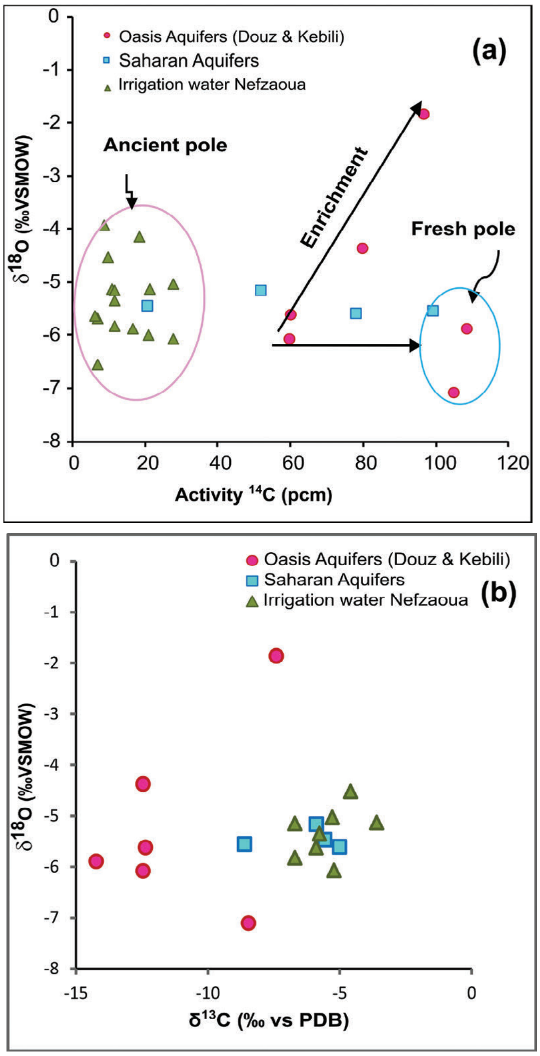

Fig. 10 - $\delta^{18}O$ contents versus ^{14}C activities (a) and $\delta^{13}C$ (vs PDB) (b) of the groundwater samples.

Fig. 10 - Contenuto di $\delta^{18}O$ vs attività ^{14}C (a) e $\delta^{13}C$ vs PDB (b) nelle acque campionate.

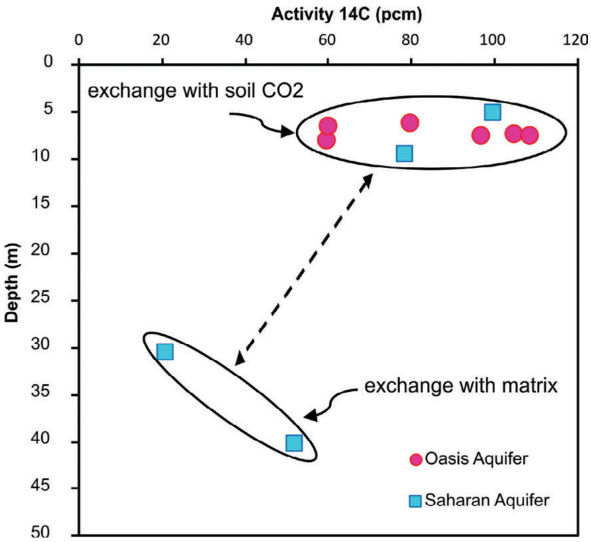

Fig. 11 - ^{14}C activities versus depth for the groundwater samples studied in the study area.

Fig. 11 - Attività ^{14}C in funzione della profondità per i campioni di acque sotterranee studiati nell'area di studio.

surface water bodies and with carbonate for the deeper samples.

The scatter plot of Fig. 11 showed the activity of different aquifers from southern Tunisia. Activity ^{14}C (pcm) versus $\delta^{13}C$ (‰ vs PDB, relative to a Pee Dee Belemnite Standard Water). Samples with more negative delta carbon-13 values are more enriched in the lighter isotope carbon-12. In contrast, points to the right are more enriched in the heavier isotope carbon-13. Points that plot in the higher carbon-14 activity are the proof of fresh water. As carbon-14 isotope decays over time, it should be expected that older water bodies will have lower carbon-14. Biogenic pole can be seen in the upper left corner of the plot, indicating a young water cluster with high activity of carbon 14. In the opposite position, (i.e., the lower right corner), mineral pole showed low carbon-14 with somewhat "higher"/less negative carbon-13. To the middle, the "mixing effect" was suggested as a possible mechanism with data points showing a trend towards ^{14}C activity increase as delta carbon-13 value decreases. To sum up, the observed data points of Figure 11 were highlighted for three main categories. It included the oasis aquifers (Douz & Kebili), saharan aquifers and irrigation water of the Nefzaoua. Water samples from the oasis aquifers (i.e., Douz & Kebili) can be seen as a tight cluster, exceeding 60 pcm, which suggests a high activity of carbon-14, but a negative delta carbon-13 value. This is somehow different from the saharan aquifer samples that plot in a wider range associated with a carbon 14 activity between 50% to 100%. As for irrigation water of the Nefzaoua area, most samples can be plotted near the "mineral pole". Such a configuration suggested carbon-14 depleted waters with slightly negative delta carbon-13 value (Fig.12). slightly negative $\delta^{13}C$ values can indicate organic matter derived from marine sources (Taylor et al., 2006). However, in the context of Chott Djerid, it is important to consider other possibilities for the organic matter source (Amaral et al., 2010).

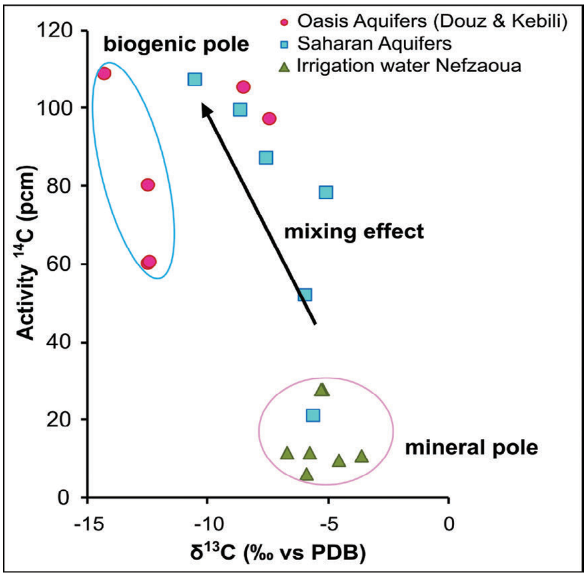

Fig. 12 - Relationship between $\delta^{13}C$ contents and ^{14}C activities.

Fig. 12 - Relazioni tra contenuto di $\delta^{13}C$ e attività ^{14}C.

Conclusions

Isotopes analysis has been applied as a powerful method to investigate the main properties of the Chott Djerid shallow aquifers. Based on the obtained results, one can recognize the following points:

- Stable isotope contents revealed distinct groups of water with:
  - A group of water resulting from recent recharge by rainwater and direct infiltration before being evaporated, formed by the waters located in the piedmont of Dahar;
  - A second group corresponded with the returning irrigation water;
  - Another group formed by the points that have had intense evaporation;
  - A group of waters (samples of the Saharan aquifers) indicated the significant contribution of the mixing effect and/or the contribution of other possible processes;
- based on tritium contents, it was concluded that recharge took place at the Dahar lowlands, reaching 13.2 mm on average;
- Deuterium excess, calculated for each water point, indicated that dissolution contributed up to 90% of the mineralization in the Nefzaoua and Djerid oasis aquifers. Our results demonstrated elevated carbon-14 activity in the oases shallow aquifers, indicating a strong influence of biogenic processes.
- Shallow aquifers of the Sahara were recharged with recent waters that have undergone mixing with the underlying deeper aquifers. Characterizing the dominant salinization processes in the Djerid area required the integration of hydrogeological, geochemical, and isotopic tools. Based on a thorough understanding of regional hydrogeology (i.e., aquifer's geological formations, groundwater flow patterns) and the presence of any potential sources of salinity (e.g., evaporite deposits, saline drainage from irrigated lands and/or water-rock interactions);
- Dissolution should be the main process of the acquisition of higher salt content, participating with a rate of 90%.

Integrating several aspects (i.e., hydrogeology, piezometry and isotopic data) enables informed water resource management by deciphering the dominant salinization process (dissolution for the oasis shallow aquifers, infiltration, mixing and evaporation for the saharan shallow aquifers).

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Author contributions

Zohra KRAIEM: Formal analysis, Writing - original draft, Writing - review & editing. Conceptualization, Methodology, Data Curation, Visualization, Validation.

Kamel ZOUARI, Najiba Chekir, Aissa Hlimi: Methodology, Visualization, Validation.

All authors have read and agreed to the published version of the manuscript.

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Competing interest

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Additional information

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