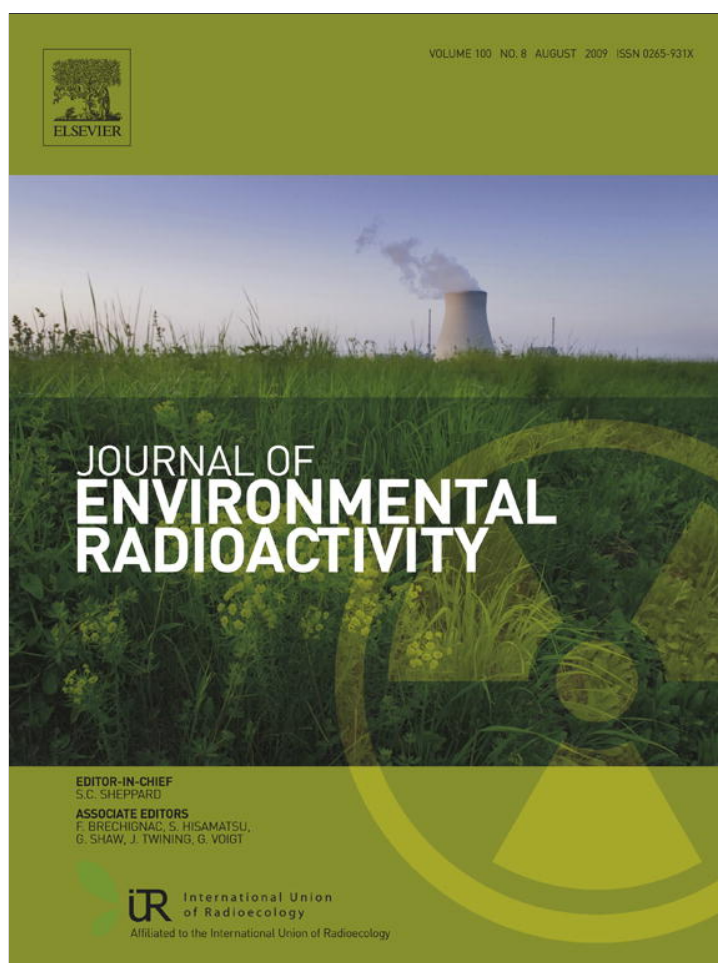




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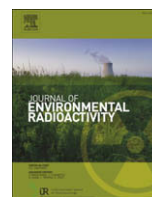
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Uranium isotopes in groundwater from the continental intercalaire aquifer in Algerian Tunisian Sahara (Northern Africa)

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ABSTRACT

The disequilibrium between ^{234}U and ^{238}U is commonly used as a tracer of groundwater flow. This paper aims to identify uranium contents and uranium isotopic disequilibrium variation in groundwater sampled from deep Continental Intercalaire aquifer (southern Algeria and Tunisia). Large variations in both U contents (0.006–3.39 ppb) and $^{234}\text{U}/^{238}\text{U}$ activity ratios (0.4–15.38) are observed. We conduct a first assessment in order to verify whether the results of our investigation support and complete previous hydrogeological and isotopic studies. The dissolved U content and $^{234}\text{U}/^{238}\text{U}$ activity ratio data were plotted on a two-dimensional diagram that was successfully utilized on sharing the CI aquifer into different compartments submitted to different oxidising/reducing conditions and leads also to distinguished two preferential flow paths in the Nefzaoua/Chott Fejej discharge area. Uranium isotopes disequilibrium indicate that uranium chemistry is mainly controlled by water–rock interaction enhanced by long residence time recognised for this aquifer.

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1. Introduction

Due to long radioactive half-lives, ^{238}U and ^{234}U isotopes are in secular equilibrium in all minerals and rocks greater than one million years old and that are closed systems for U. The $^{234}\text{U}/^{238}\text{U}$ activity ratio (AR) is therefore unity in the bulk of such systems. In natural waters, radionuclides are carried via normal recharge (cosmogenic nuclides) and as a result of water–rock interaction (natural decay series nuclides). The ratio of ^{234}U to ^{238}U is almost always found to differ significantly from its equilibrium value. These isotopic disequilibria result from geochemical as well as physical conditions (Osmond and Cowart, 1992, 1976; Ivanovich and Harmon, 1982). Aquifers environments are systems where water and minerals are strongly mixed and thus where both chemical and physical differentiation processes are particularly enhanced according to rock–water ratio, surface area and residence time. These conditions are often observed on deep and fossil aquifers. Various mechanisms have been suggested to interpret the generation of elevated AR's in solution. For instance, Rosholt et al., (1963) proposed the occurrence of enhanced chemical solution of

^{234}U due to radiation damage to crystal lattices or to autoxidation from U^{4+} to U^{6+} resulting from the decay of the parent ^{238}U .

The application of uranium isotopic disequilibrium to solve hydrogeological problems is not as well spread as stable isotope of the water molecule. However, in several studies (Osmond et al., 1968; Meyer, 1989; Banner et al., 1991; Roback et al., 1997; Suski et al., 2001; Dabous and Osmond, 2001; Suksi et al., 2006; Bonotto, 2006), uranium series disequilibria successfully help in the description of aquifer processes. The main result of these investigations was to classify aquifers on steady state, augmenting or decaying systems in terms of dissolved uranium content value and in term of $^{234}\text{U}/^{238}\text{U}$ activity ratio values. Specific data analyses also help to refine groundwater mixing processes (Suski et al., 2001; Suksi et al., 2006).

In this article, data of U contents and isotopic activity ratio in groundwater sampled from Continental Intercalaire aquifer, a fossil complex huge aquifer system situated in the northern Sahara, are described for 30 sites distributed along main flow paths from recharge areas (Algeria basin) to discharge areas (Tunisian basin). Uranium isotope signatures are interpreted along the defined groundwater transport pathway in an attempt to identify main uranium chemistry mechanisms.

We have adopted a two-step methodology. First, we attempt to characterize groundwater aquifer in terms of U-contents: oxidized

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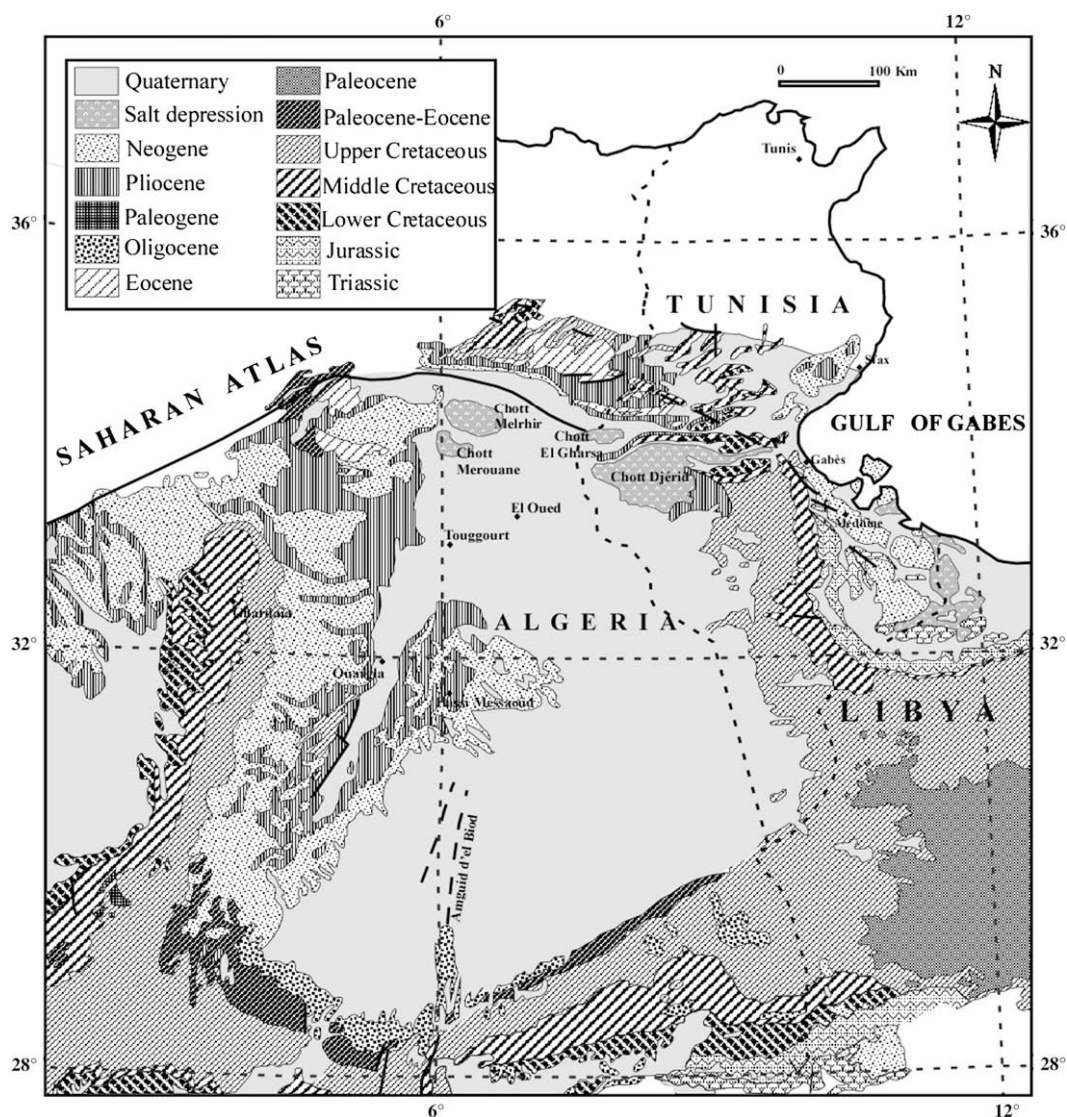


Fig. 1. Location and geological features of the Saharan sedimentary basin in North Africa (after OSS, 2003).

aquifer on “normal” U-content strata (values between 1 and 10 ppb); oxidized aquifer on enhanced U-content strata (values higher than 10 ppb); reduced aquifer on low U-content strata (values lower than 1 ppb) and in terms of $^{234}\text{U}/^{238}\text{U}$ activity ratios data: “normal” world-wide groundwater (values between 1 and 2), possibility of formation processes (values higher than 2) or remobilization (values lower than 1). Then, we attempt to delineate preferential flow lines across the aquifer based on a two-dimensional diagram of reciprocal uranium vs uranium isotopes activity ratios.

2. Site description

The *Continental Intercalaire* (CI) aquifer underlies all the Bas-Sahara in an arid region where rainfall amounts average around 100 mm/y. The huge groundwater reservoir of the CI is one of world largest aquifer system, with a surface of 600 000 km². This aquifer is lodged in lower Cretaceous continental formations (Neocomian, Barremian, Aptian and Albian) which thickness and lithology show significant lateral variation (ERESS, 1972; Confiantini et al., 1974; Aranyosy and Mamou, 1986; Mamou, 1990; Edmunds et al., 1997; Gries, 2000). The aquifer is however continuous over the whole

basin from north to south from the Saharan Atlas to the Tassili mountains of the Hoggar, and west to east from western Algeria to the Libyan desert. A major N–S structure, the M'zab dorsal (Cornet, 1964; ERESS, 1972; Bishop, 1975) divides the basin into two sub-basins (occidental and oriental). The studied area is situated in the oriental basin (Great Oriental Erg), which crosses Algeria and Tunisia (Fig. 1). It's limited on the north by the Saharan Atlas mountains (900 to 1100 m), on the west by the M'zab dorsal, on the south by the Tinher plateau, and on the east by the uplands of the Dahar mountains (500–700 m).

The aquifer roof becomes deeper from south to north. The minimum depth (0–500 m) is observed along the occidental border of the Dahar mountain and near the Tademaït plateau while the maximum (1500–3500 m depth) is mainly located in the region of the endoreic closed depressions of Chott Melrhir (Algeria) and Chott Djérid (Tunisia), giving rise to a high artesian pressure.

Principal geological features in Algeria and Tunisia (Fig. 2) show important spatial variations. In the centre of the basin, the aquifer is hosted in purely continental formations; while first lacustrine and marine facies appear towards the borders, especially in the east. The aquifer thickness varies between 100 m in the recharge area in the western part of the basin region up to more than 1500 m in the

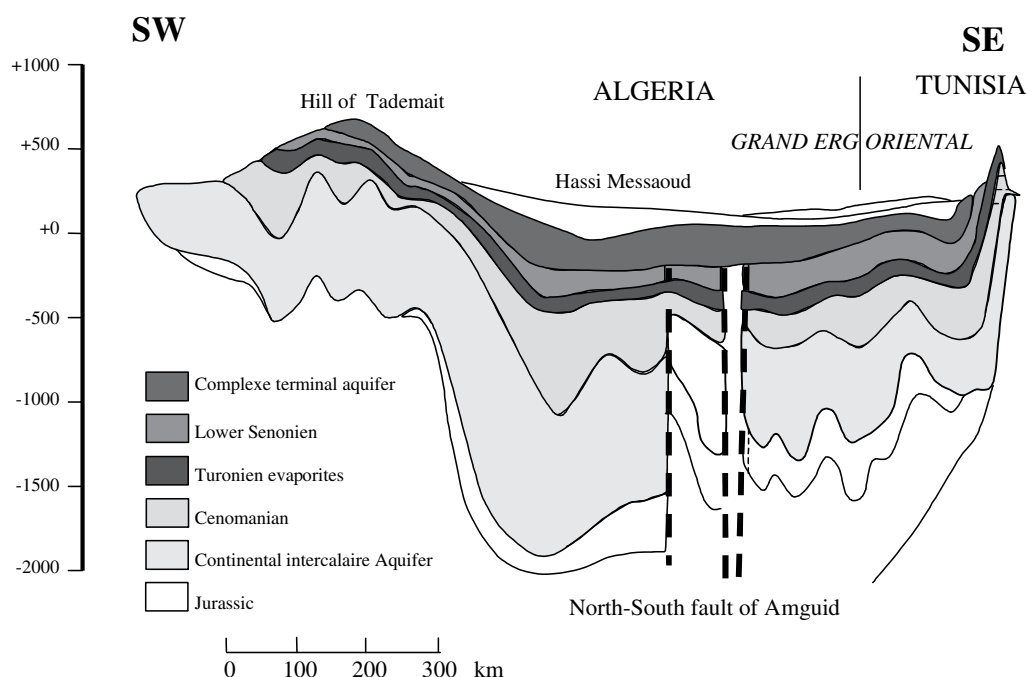


Fig. 2. Schematic section through the Continental Intercalaire aquifer through Algerian Tunisian basins (after Edmunds et al., 1997).

Djérid region. A high degree of heterogeneity is observed in the heart of the CI aquifer due to the succession of different detrital sediments formations separated by clay-rich strata. Some thin but relatively permeable horizons may be very important in controlling groundwater flow and hydraulic continuity. This phenomenon is particularly marked where the Saharan platform passes to the lower-Sahara region in the south: thickening of the units and lithological changes give rise to changes in piezometry and chemical composition of water. Tectonic activities and thus faulting has played a major role in the compartmentalisation of the southern (El Amguid fault) and in the eastern (Elhamma fault) parts of the CI aquifer (ERESS, 1972; Edmunds et al., 1997).

Groundwater flows in the CI of the Great Oriental Erg converge towards a single discharge zone in Tunisia (Chott Djérid/Chott Fejej region and Gulf of Gabès). The main W-E flow path, comes from the Saharan Atlas eastward the Chott Djérid and the gulf of Gabès in Tunisia. Two secondary flow directions may be distinguished: SW-NE from the M'zab dorsal southwestern Algeria eastward the chott Fejej/Gulf of Gabès in Tunisia and S-N from the Dahar region of Tunisia (Fig. 3). Hence, two major regions have to be distinguished in the Tunisian basin of the CI aquifer: In the Djérid region, the aquifer is mainly lodged in continental formations extending from the Neocomian at the base to the Albian and the hydraulic continuity seems to be only ensured by the west-east flow component across the Algerian–Tunisian border (Mamou, 1986; Edmunds et al., 1997). On the other hand, in the Nefzaou–Chott Fejej region, the aquifer is hosted in thick deltaic terrigenous sediments (Mamou, 1990) and groundwater flows along a north-north east line coming from a recharge (or paleorecharge) area located in the M'zab dorsal southwestern Algeria and in the plateau of Tihert in southern Algeria and a second recharge area located in the Dahar uplands in southern Tunisia (Edmunds et al., 1997).

3. Material and method

Groundwater samples were collected from boreholes along main flow paths of the CI aquifer over the Algerian and the Tunisian basins (Fig. 3). The interval from which water is derived, are between 100 m in the recharge area to more than

2500 m in the deepest part of the aquifer in the area of the Tunisian chotts. Groundwater temperature increases from less than 20 °C in the recharge area to above 70 °C in the deep part of the basin. The conductivity, as a measure of the total mineralization, increases from around 1500 to 6000 $\mu\text{S cm}^{-1}$ across the aquifer but is influenced by different sources of salinity and different redox conditions in the aquifer (Edmunds et al., 1997; Guendouz, 1985).

Groundwater for uranium isotopes analyses have been collected (20 l) from 30 wells (12 across the algerian basin and 17 from the Tunisian basin) during different sampling campaigns. Samples are filtered through 0.45 μm and spiked with ^{232}U tracer for radiochemical yield; 6 ml of a FeCl_3 solution (0.35 g/l) was added, and U was co-precipitated with $\text{Fe}(\text{OH})_3$ using NH_4OH solution. The precipitate is isolated by centrifugation and then dissolved by chlorhydric acid (8 N) to be isolated by ion exchange using Dowex AG-1X8 resin and purified by solvent extraction techniques (Tomé and Sanchez, 1990). Uranium is then electroplated onto a stainless-steel disk that allowed α -counting. ^{234}U and ^{238}U activities are counted by α -spectrometry as much time as necessary to reach a statistical error below 10%. Table 1 shows the activity of ^{234}U and ^{238}U , $^{234}\text{U}/^{238}\text{U}$ activity ratio, field information (location, capture depth), salinity and physical data (pH, temperature) for all samples.

4. Results and discussion

Uranium concentrations in groundwater from the CI aquifer range widely from 0.006 to 3.39 ppb while $^{234}\text{U}/^{238}\text{U}$ activity ratios vary between 0.4 and 15.38 (Table 2).

Uranium contents as well as of $^{234}\text{U}/^{238}\text{U}$ AR do not vary systematically across the study area but some trends can be observed on U-contents vs $^{234}\text{U}/^{238}\text{U}$ AR diagram (Fig. 4).

Groundwater can be classified according two parameters: uranium concentration and $^{234}\text{U}/^{238}\text{U}$ activity ratio (Coward and Osmond, 1980; Osmond and Cowart, 1983; Toulhoat et al., 1988; Bonotto, 1999). U-content values allow to classify aquifers in oxidized aquifer on “normal” U-content strata (values between 1 and 10 ppb), oxidized aquifer on enhanced U-content strata (values higher than 10 ppb) and reduced aquifer on low U-content strata (values lower than 1 ppb) whilst $^{234}\text{U}/^{238}\text{U}$ activity ratios distinguish between “normal” world-wide groundwater (values between 1 and 2); possible U-accumulation (values higher than 2), or possible remobilization of a uranium (values lower than 1). High activity ratios (>2) can be due to a higher than normal ratio of leachable uranium in aquifer or amorphous uraninite, enhancing

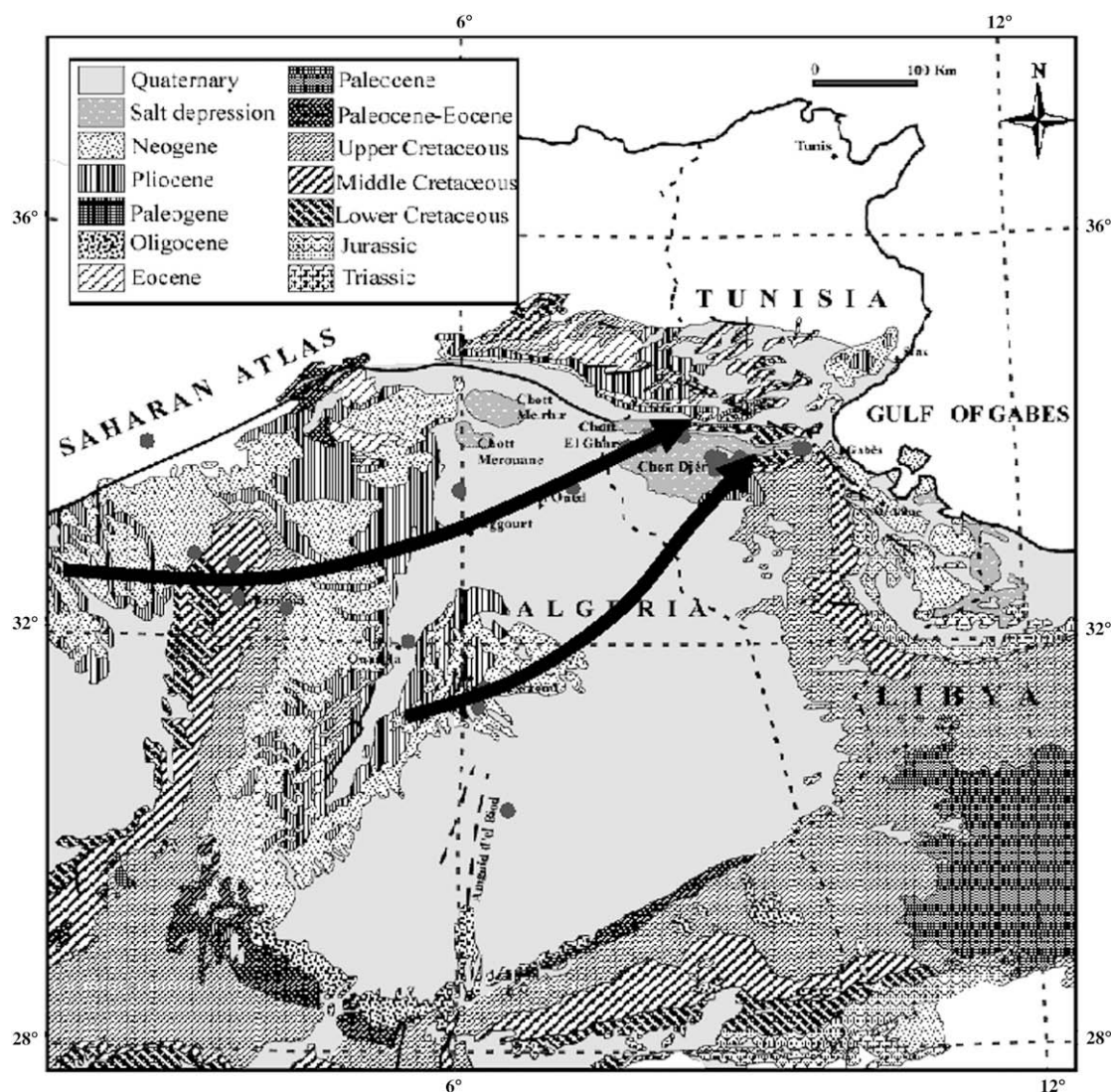


Fig. 3. Main flow lines within CI aquifer from the recharge zone of the Atlas Mountains to the discharge area in the Gulf of Gabès – Samples location.

α -recoil. An activity ratio lower than 1 implies intense dissolution. Uranium concentration levels reflect both redox conditions and uranium concentrations in surrounding rocks (Coward and Osmond, 1980; Bonotto and Andrews, 2000; Roback et al., 1997). In oxidizing conditions, uranium is very soluble as U(VI)-complexes, mainly carbonates. In reducing conditions, uranium is insoluble as U(IV)-oxides.

According to this classification, the U content and AR measured in the CI aquifer are greatly variable, as shown in Fig. 4 where samples are fitted in practically all fields. However, along the main flow crossing the basin from west (Saharan Atlas in Algeria) to east (Djérid region in Tunisia), the CI aquifer can be shared in an upstream oxidized compartment and a downstream reduced compartment (Fig. 4). The spatial distribution of groundwater data reveals that the oxidized portion, with relatively high uranium concentration (1.78–3.39 ppb) and moderate $^{234}\text{U}/^{238}\text{U}$ activity ratios (2.05–4.27) is restricted on the northwestern area of the survey zone. It is limited by a boundary located between $4^{\circ}30'\text{E}$ and 5°E of longitude (Fig. 5) and corresponding to the redox front previously identified by spatial relationships of several redox sensitive elements such as Fe, Mn or Cr (Guendouz, 1985; Edmunds et al., 1997). This compartment can be considered as a “steady-state” aquifer: oxidized aquifer on “normal” U-content strata

(U-contents values ranging between 1 and 10 ppb) with possible U-accumulation processes ($\text{ARs} > 2$) but not necessarily of economic size and grade. In this area, $^{234}\text{U}/^{238}\text{U}$ ARs are broadly decreasing along the main W-E flow line from recharge area situated on the southern side of Saharan Atlas toward the redox boundary. The only anomaly observed is for the “Hadjira” sample but it could be explained by a deeper capture depth (1900 m) compared with other samples of this upstream compartment for which sampling depth is increasing from the recharge area toward the redox boundary appearing at appreciatively 1000 m depth.

The distribution of uranium content and uranium isotopic disequilibrium in the reduced part of the aquifer is less homogeneous and is highly influenced by lithostratigraphic conditions (Chkir and Zouari, 2007). Uranium concentrations range between 0.006 and 0.52 ppb while $^{234}\text{U}/^{238}\text{U}$ AR range between 1.49 and 15.38 in the Djerid region. Hence, this second part of the CI aquifer in the Djerid region can be defined as a reduced aquifer with possible remobilization of uranium or enhanced α -recoil processes. Uranium concentrations remain very low along the main flow line while $^{234}\text{U}/^{238}\text{U}$ ARs reach very high values (2 or 3 magnitude of those of the upstream oxidized part of the aquifer).

Such high $^{234}\text{U}/^{238}\text{U}$ ARs have already been found in other flow systems. In a slow-moving relatively stable system, high uranium

Table 1

Uranium concentration and isotopic ratios.

No	Nom	Latitude (°N)	Longitude (°E)	U ppb	2σ	²³⁴ U/ ²³⁸ U	2σ	Depth, m	RS, mg/l	Temp. (°C)	pH
1	Berriane 2	32.771	3.670	2.87	0.01	3.19	0.02	545	964	28.6	7.56
2	CedadaCl	34.012	8.257	0.025	0.003	11.02	0.86	2371	2260	50	7.67
3	CFF3	33.895	9.632	2.354	0.238	1.65	0.231	950	2770	63.50	6.67
4	CFF9	33.890	9.688	1.539	0.161	1.71	0.245	886	3420	63.50	6.72
5	ChorfaCl4	33.788	8.833	0.056	0.005	7.84	0.79	2190	2210	59.5	6.8
6	DebabchaCl4	33.802	8.736	0.054	0.005	8.84	0.94	2295	2030	68.1	6.7
7	Djamâa	33.481	5.994	0.031	0.001	0.4	0.22	1755	1719	46.5	7.82
8	ElHammaCl1Bis	33.989	8.193	0.12	0.001	5.7	0.67	1550	2750	69.8	7.42
9	ElHammaCl4	34.007	8.446	0.015	0.005	8.37	1.33	1475	2720	69.5	7.4
10	El-Oued (Seh.Bery)	33.508	7.169	0.52	0.03	1.496	0.086	850	2449	35.9	7.8
11	Gassi-touil GT110	30.376	6.501	0.034	0.001	2.53	0.09	955	4064	43.9	7.26
12	Ghardaia (Bouhra)	32.432	3.717	2.86	0.3	3.39	0.169	390	1618	29.1	7.23
13	Ghardaia (D. Bend)	32.545	3.595	2.89	0.14	3.66	0.02	467	1534	31.6	7.41
14	Hadjira	32.881	5.264	1.78	0.014	2.05	0.02	1895	1767	50.8	7.6
15	Hassi mess MDH101	31.555	6.256	0.034	0.001	2.53	0.09	1300	1687	52.8	7.64
16	HazouaCl	33.900	7.872	0.008	0.002	9.92	1.37	2059	1800	59.1	7.24
17	KebiliCl6	33.683	8.770	0.483	0.018	3.97	0.16	2774	2220	63.7	7.3
18	Laghouat (Assafia)	33.961	2.781	2.99	0.13	4.265	0.087	100	1181	20.6	7.32
19	LimagueusCl8	33.779	8.752	0.259	0.102	4.89	0.2	1730	2420	67	7.0
20	MahacenCl1	34.016	8.242	0.089	0.001	5.55	0.76	2557	2240	63.8	7.31
21	MansouraCl3	33.727	8.943	0.044	0.003	7.94	0.58	2557	3000	62.9	7.3
22	NeftaCl2	33.886	7.877	0.006	0.001	15.38	1.32	2584	2670	66.7	7.31
23	NeftaCl3	33.886	7.877	0.013	0.003	12.94	1.27	2190	2820	57.9	7.68
24	Ouargla (Hadeb)	32.019	5.469	0.22	0.003	1.75	0.029	1450	1745	48.5	7.58
25	Rhoude El baguel	31.365	6.970	0.01	0.0005	3.17	0.19	995	2713	40	7.42
26	SeftimiCl7	33.802	9.017	0.568	0.016	3.19	0.1	1786	2440	67.6	6.9
27	TazraritCl	33.917	8.112	0.017	0.001	4.4	0.38	2186	4120	52.4	7.38
28	TozeurCl4	34.005	8.089	0.009	0.002	7.9	1.03	1981	4880	54.4	7.44
29	Zelfana 2	32.334	4.216	3.39	0.019	2.71	0.018	1000	1631	37.8	7.59

concentrations are seldom found, this statement emphasizes that once precipitated at the barrier front of a permanent aquifer, uranium remains in place in the solid state for a long term (Osmond and Cowart, 1992), the same is not true for ²³⁴U. If conditions at the front are near chemical equilibrium, the effects of decay and recoil may be enough to cause preferential mobilization of the daughter. Therefore, high ²³⁴U/²³⁸U AR may be found in the groundwater near such stable front and also for some distance downflow. The size, climate, lithology, stratigraphy, hydrogeology, geochemical conditions, and extent of rock–water interactions are also factors responsible of the high variability of AR values in groundwater (Bonotto and Andrews, 2000; Bonotto, 1999). Moreover, alternative mechanisms have been suggested for the generation of enhanced ²³⁴U/²³⁸U AR ratios including preferential chemical dissolution of ²³⁴U (Rosholt et al., 1963) and daughter recoil into solution by the alpha-recoil at the rock–water interface (Kigoshi, 1971). These mechanisms are favoured (Osmond and Cowart, 1992; Ivanovich et al., 1991) by long period of interaction between water of low dissolved uranium contents and hosting formations of relatively higher uranium concentrations.

Several studies have reported activity ratios reaching 12 in large confined aquifers with steady long term system such as 9.0 for deeper and older shallow groundwater of the Transvaal Dolomite Aquifer in South Africa (Kronfeld et al., 1994), 10.1 for groundwater sampled from a deep well in Ramallah (Kronfeld et al., 1975), 10.5 in confined aquifers in the Cambrian and Ordovician bedrock in the tri-state region of Missouri, Kansas and Oklahoma (Cowart and Osmond, 1980), 11.8 at a redox front of the Milk River aquifer

(Ivanovich et al., 1991) and 12.3 in a confined sandstone aquifer in Texas (Kronfeld, 1974).

Activity ratios as high as 28 have also been observed in the Brazilian part of the Guarani aquifer (Bonotto, 2006) and have been explained by the large extension of the system enhancing the three main factors responsible of uranium isotopes disequilibria (rock–water interactions, preferential chemical dissolution and alpha-recoil release of ²³⁴U).

On the other hand, ²³⁴U/²³⁸U activity ratios for groundwater samples collected from the Cambrian and Ordovician (USA) range from 2.0 in the central part of the aquifers to 40.7 in the south. The lowest ratios occur in primary recharge zones in regions where the Cambrian and Ordovician are unconfined. Activity ratios increase along flow path and with residence time of groundwater and ratios ranging from 20 to 25 reflect the location of a zone of groundwater discharge with a component coming from regions where the

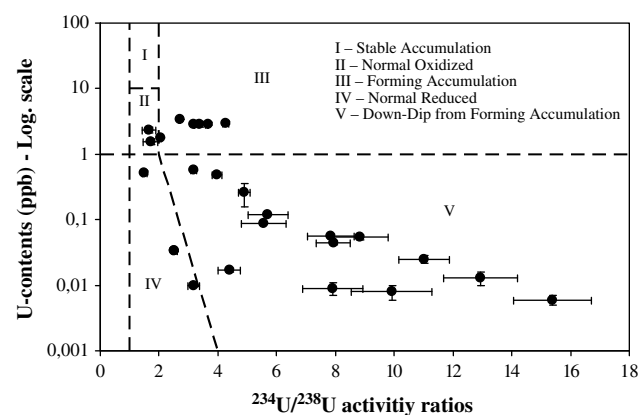


Fig. 4. The data for dissolved uranium in groundwater from Algerian Tunisian basin of the CI aquifer plotted on the U concentration vs ²³⁴U/²³⁸U activity ratio diagram proposed by Cowart and Osmond, (1980).

Table 2

Mean values, standard deviations and ranges of uranium concentrations and uranium isotopes activity ratios in the investigated samples.

	Mean value	Standard deviation	Range
U-contents (ppb)	0.877	1.174	0.006–3.39
²³⁴ U/ ²³⁸ U	5.023	3.714	0.4–15.38

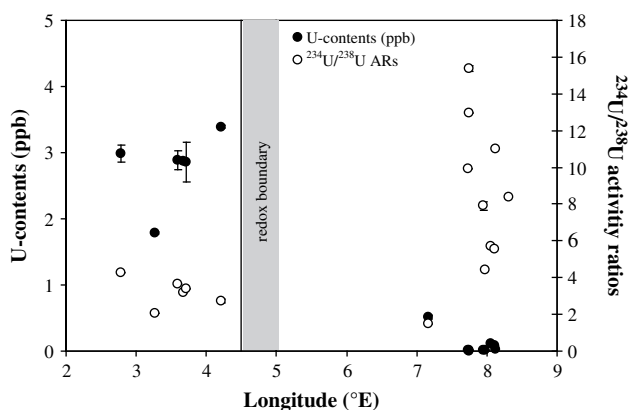


Fig. 5. Uranium content and $^{234}\text{U}/^{238}\text{U}$ activity ratio along the main W-E flow path crossing the Grand Erg Oriental from West (Algeria) to East (Tunisia).

Cambrian and Ordovician is confined (Gilkerson et al., 1984). The highest activity ratios (values from 25 to 40) occur in the eastern part of the Cambrian and Ordovician aquifers where groundwater would have been recharged during the Wisconsin. So high activity ratios are not unique since $^{234}\text{U}/^{238}\text{U}$ ARs reported for the continental intercalaire aquifer of the Irhazer Plain in Niger (Andrews et al., 1994) increase by preferential ^{234}U from 1.3–3.4 in the outcrop zone to 42 in the south-western confined zone close to U deposition zone or just downstream of it.

The two later cases are quite uncommon and reflect local and specific geological settings. Nowadays, 28 is a maximum value of uranium isotopes disequilibrium that could be explained by hydro-geochemical settings of flow systems.

In the reduced part of the CI aquifer, along the main W-E flow line, the reducing conditions become dominant after redox boundary inducing an U-precipitation and groundwater with low dissolved uranium contents. However, even if uranium chemistry in this area remains quite stable, the increase of temperature (up to 70 °C) are favourable to promote U dissolution at the greater depths of the CI aquifer. Since the α -recoil effect enhances ^{234}U dissolution, it may be thought that the long residence time of groundwater in this area leads to the highest uranium activity ratios observed downflow the aquifer.

The same data treatment has been applied along the secondary SW-NE flow line from the M'zab dorsal to the Nefzaou/chott Fejej area (Fig. 3). The very low uranium concentrations are typically indicating reducing conditions in the confined aquifer beneath the Oriental Grand Erg in southern Tunisia and Algeria. Higher concentrations in the discharge area in the Nefzaou/Chott Fejej region could be related to different redox conditions or complexation. In order to highlight the different processes in the aquifer, a preliminary interpretation of these data is attempted using the diagram relating reciprocal U-content vs the $^{234}\text{U}/^{238}\text{U}$ AR (Osmond and Cowart, 1983, 1992). In this diagram, $^{234}\text{U}/^{238}\text{U}$ ARs plotted against reciprocal U concentrations on the mixing diagrams exhibit a linear trend extending from the lower left (low AR with high concentrations) to upper right (high AR with low concentrations). This can be interpreted as mixing lines (Dabous and Osmond, 2001; Chkir and Zouari, 2007); at one end is very dilute aquifer water, which has experienced little leaching of U (upper right), and at the other end member is the concentrated leachate of the host rock (lower left). The dispersion of points between these two end members indicates water with various mixture ratios. The trend line is defined by two parameters: the y-intercept, which is the $^{234}\text{U}/^{238}\text{U}$ AR of the U being leached from the host rock, and the slope of the trend line, which is the ^{234}U -excess per kilogram

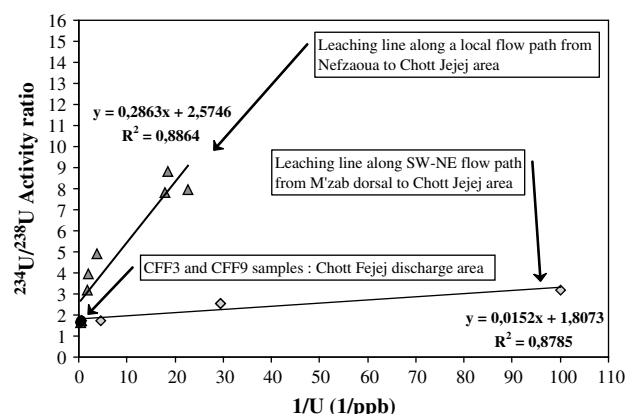


Fig. 6. Reciprocal uranium content vs uranium isotopes activity ratios for the secondary SW-NE flow path.

(U-equivalent ppb) and is characteristic of the pre-leach water. ^{234}U -excess corresponds to the hypothetical concentration that the ^{234}U isotope has in excess to the ^{238}U concentration and has no real physical meaning as the actual mass of the ^{234}U isotope is $\sim 18\,000$ times less than the ^{238}U mass (Cizdziel et al., 2005). The unit for ^{234}U -excess is U-equivalent parts per billion, which is the concentration in micrograms per kilogram of an equilibrated amount of ^{238}U (^{234}U activity per mass of water) (Dabous and Osmond, 2001). However, the interpretation of the data has to be supported by the hydrogeological and hydrochemical characteristics of aquifers.

Along the secondary SW-NE flow line, two mixing lines flowing from very dilute aquifer water with low dissolved U (upper right), to concentrated leachate of the host rock (lower left) appear to characterize the CI groundwater (Fig. 6). These two lines have different parameters but converge toward two samples from the Chott Fejej discharge area indicating that groundwater in this area have different origins. The first line extends from the M'zab dorsal recharge or paleorecharge area with a ^{234}U -excess of about 0.015 (U-equivalent ppb) and the AR of the host rock U is 1.82. The second mixing line seems to be a more local flow line from the Nefzaoua area toward the same discharge area of the Chott Fejej. This line is also characteristic of a dissolution process with a ^{234}U -excess of about 0.27 (U-equivalent ppb) and the AR of the host rock U is 2.57. This line has already been identified (Chkir and Zouari, 2007) and is refined here by two points (CF3 and CF9) sampled more recently in the discharge area.

The first end member (upper right), which is expected to be very dilute aquifer water, corresponds to water sampled from the most confined and the deepest part of the aquifer as observed by Dabous and Osmond (2001) for Nubian aquifer (Egypt). Low uranium contents could be explained by high reducing conditions enhancing uranium precipitation. The second end member (lower left) which is expected to be concentrated leachate of the host rock corresponds to water sampled in the discharge area of chott Fejej.

The increase of dissolved uranium along these flow line indicates that the leaching process is predominant compared to alpha-recoil effect hence activity ratios decrease simultaneously. Thus, the water-exchange is the dominant mechanism that controls groundwater uranium chemistry as for other chemical elements (Edmunds et al., 1997, 2003).

A previous study applied to uranium contents of groundwater sampled in the chott Djérid region has risen that uranium chemistry is mainly characterised by leaching process with preferential ^{234}U dissolution enhanced by a weak hydraulic gradient facilitating the water–rock interaction (Chkir and Zouari, 2007). For the Djérid

area, ^{234}U -excess is about 0.045 (U-equivalent ppb) and the AR of the host rock U is 5.58. The two points recently sampled (CF3 and CF9) have not been included in this leaching line because no hydrodynamic continuity can be proved. Moreover, this leaching line has not been back extended to the recharge zone because of data lack. In fact, this zone is an extremely remote area with difficult access and the sampling of groundwater has been possible mainly by using water from wells used for oil exploration and not all sampled water have been analysed for uranium investigation.

5. Conclusion

This study was realized to investigate potential use of uranium isotopic variation for characterizing one of the greatest fossil groundwater in world, located over the Sahara, from Western Atlas Chain in western Algeria to Eastern North Africa along Mediterranean coast Tunisia and to southern to the Tassili of the Hoggar. We intend to use long-life radionuclide (^{234}U and ^{238}U) in order to identify different categories of groundwater along the main W-E and the secondary SW-NE flow lines across Algerian and Tunisian basins.

The size, climate, lithology, stratigraphy, hydrogeology, geochemical conditions, and extent of rock–water interactions as well as alternative mechanisms related to radioactive phenomenon are among the factors responsible of the very variable uranium isotopes disequilibrium values in such huge and complex aquifer. The Continental Intercalaire aquifer has a wide range of uranium contents (0.006–3.39 ppb) decreasing from the recharge area on the west (Algerian Atlas) to the discharge area of the east (Chott Djérid/Gulf of Gabès) and very scattered $^{234}\text{U}/^{238}\text{U}$ activity ratios (0.4–15.4) decreasing along this main W-E flow path. $^{234}\text{U}/^{238}\text{U}$ activity ratio have already been reported for deep groundwater and have been explained either by a long history of high surface area interaction between a water of low uranium solubility and an aquifer rock with relatively higher concentration of uranium (factor of 1000) (Kronfeld et al., 1975) or by an elevated temperatures that may cause rock–water isotopic re-equilibration to occur (Kramer and Kharaka, 1986). High $^{234}\text{U}/^{238}\text{U}$ activity ratios are often observed in large confined aquifer with steady long-term flow systems such Continental Intercalaire aquifer.

The drop of uranium content values along the main W-E flow line is abrupt and allows to localize between $4^{\circ}30'\text{E}$ and 5°E of longitude the redox front that shares the CI aquifer on a oxidized compartment on its western part and a reduced compartment on its eastern part. Uranium contents and uranium isotopic disequilibrium values and variation in the oxidized compartment is characteristic of a “steady-state” aquifer lodged on enhanced uranium content strata and submitted to the decay of ^{234}U -excess along flow lines. Whilst, the eastern part of the CI aquifer in the Djérid region has been found to be a reduced aquifer where uranium chemistry is controlled by possible remobilization or enhanced α -recoil effect. On the other hand, different processes have been identified along the second flow line indicating different mechanisms. Indeed, more detailed investigations using reciprocal uranium vs activity ratios diagram have lead to distinguish two different origins for groundwater sampled in the Chott Fejej discharge area: the first preferential flow path is carrying groundwater from the recharge area situated in the M'zab dorsal in Southern Algeria while the second preferential flow path is thought to be a local flow line crossing the Nefzaoua region toward the Chott Fejej discharge one. The main mechanism controlling uranium chemistry along these two flow lines appears to be uranium leaching from the hosting formations enhanced by long residence time favourite by extended water–rock interactions.

The present study has, also, pointed out areas which need further studies, in order to better understand the variation of uranium concentration in groundwater related to geochemical and hydrodynamical processes.

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