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1 **Uranium isotopes as tracers of groundwater evolution in the Complexe**
2 **Terminal aquifer of southern Tunisia**

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22 **Abstract**

23 The Complexe Terminal (CT) multi-layer aquifer is formed by Neogene/Paleogene sand
24 deposits, Upper Senonian (Campanian-Maastrichtian limestones) and Turonian carbonates.
25 The chemical composition and isotopes of carbon and uranium were investigated in
26 groundwater sampled from the main hydrogeological units of the (CT) aquifer in southern
27 Tunisia. We paid special attention to the variability of uranium contents and isotopes ratio
28 ($^{234}\text{U}/^{238}\text{U}$) to provide a better understanding of the evolution of the groundwater system.
29 Uranium concentrations range from 1.5 to 19.5 ppb, typical of oxic or mildly reducing
30 conditions in groundwaters. The lowest concentrations are found southeast of the study area,
31 where active recharge is supposed to take place.

32 When looking at the isotope composition, it appears that all the samples, including those from
33 carbonate levels, are in radioactive disequilibrium with significant ^{234}U excess. A clear-cut
34 distinction is observed between Turonian and Senonian carbonate aquifers on the one hand,
35 with $^{234}\text{U}/^{238}\text{U}$ activity ratios between 1.1 and 1.8, and the sandy aquifer on the other hand,
36 showing higher ratios from 1.8 to 3.2. The distribution of uranium in this complex aquifer
37 system seems to be in agreement with the lithological variability and are ultimately a function
38 of a number of physical and chemical factors including the uranium content of the hosting
39 geological formation, water-rock interaction and mixing between waters having different
40 isotopic signatures. Significant relationships also appear when comparing the uranium
41 distribution with the major ions composition. It is noticeable that uranium is better correlated
42 with sulfate, calcium and magnesium than with other major ions as chloride or bicarbonate.

43 The ^{14}C activities and $\delta^{13}\text{C}$ values of DIC cover a wide range of values, from 1.1 pmc to 30.2
44 pmc and from -3.6‰ to -10.7‰, respectively. ^{14}C model ages estimated by the Fontes and
45 Garnier model are all younger than 22 Ka and indicate that the recharge of CT groundwater
46 occurred mainly during the end of the last Glacial and throughout the Holocene.

47 **Keywords:** CT Southern Tunisia; Uranium isotopes; Radicarbon; Holocene; water-rock
48 interaction; mixing.

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55 **1. Introduction**

56 Identification of the parameters controlling the uranium concentration and the origin of the
57 $^{234}\text{U}/^{238}\text{U}$ fractionation is essential for the potential use of ($^{234}\text{U}/^{238}\text{U}$) activity ratios as a tracer
58 in hydrogeological investigations. Uranium exists in two oxidation states (U^{IV} or U^{VI}) in
59 groundwater, depending on the environmental conditions. U^{4+} forms insoluble reduced
60 chemical species, while U^{6+} is highly soluble in oxidizing natural waters, occurring as the
61 uranyl cation, and tends to form stable carbonate complexes such as $\text{UO}_2(\text{CO}_3)_2^{2-}$ (McKelvey
62 et al., 1955; Langmuir, D., 1978). The concentration and the mobility of uranium isotopes are
63 function of a number of physical and chemical factors including the uranium content of the
64 aquifer rocks and its mineralogical composition, the path lengths and contact time between
65 the aquifer matrix and the flowing water, water-rock interaction and mixing between waters
66 having different isotopic signatures (Riotte et al., 2003; Durand et al., 2005; Maher et al.,
67 2006; Elliot et al., 2014).

68 Improving our understanding of the interplay of the different factors controlling uranium
69 geochemistry is thus the main topic of this study, carried out on the scale of a large and
70 complex hydrogeological system. The “Complexe Terminal” (CT) aquifer of the Chott basin
71 in southern Tunisia constitutes the main groundwater resource in the study area, characterized
72 by an arid/semi-arid climate with irregular rainfall and very high evapotranspiration rates. In
73 spite of these climatic conditions, oasis agriculture activities based on irrigation have been
74 widely expanded since the second half of the 20th century, strongly increasing the pressure on
75 water resources. Nowadays, the overexploitation of this aquifer system is revealed by spring
76 drying, decreasing piezometric heads, and degradation in water quality (OSS, 2003;
77 Zammouri et al, 2007; Hadj Ammar et al., 2014). Therefore, the ability to use fossil
78 groundwater to buffer arid climate and rainfall deficits is now heavily constrained by the need
79 to protect the groundwater systems. In the current context of climate change and increasing

80 demand for water, it is essential to understand the groundwater origin, flow dynamics and
81 mean residence times for the assessment of reliability of these resources as a major water
82 supply. A large number of studies have already been devoted to the questions of groundwater
83 residence time and recharge rate over Northern Africa (Guendouz et al., 1997; Edmunds et al.,
84 2004; Baba Sy, 2005; Chkir and Zouari, 2007; Abid et al., 2009; Abid et al., 2010; Hadj
85 Ammar et al., 2010; Abid et al., 2011; Abid et al., 2012; Moulla et al., 2012; Gonçalves et al.
86 2013; Hadj Ammar et al., 2014; Petersen et al., 2014; Petersen et al., 2018). However, the
87 precise reconstruction of the groundwater response to climate changes largely remains an
88 open question in this region, still based on limited data sets and models.

89 Numerous studies have demonstrated the potential use of the $^{234}\text{U}/^{238}\text{U}$ activity ratio as a
90 tracer of water rock interactions and evolution of groundwater chemistry (Osmond et al.,
91 1974; Andrews et al., 1982; Andrews and Kay, 1983; Pearson et al., 1983; Frohlich and
92 Gellermann, 1987; Ivanovitch et al., 1991; Riotte and Chabaux, 1999; Riotte et al., 2003;
93 Frohlich, 2013; Paces and Wurster, 2014; Méjean et al., 2016), although its use to determine
94 water residence times is generally hampered by the complexities of ^{234}U fractionation. In this
95 respect, several studies have been focused previously on southern Tunisia (Chkir and Zouari,
96 2007; Chkir et al., 2009; Hadj Ammar et al., 2010; Chkir et al., 2012; Dhaoui et al., 2016),
97 providing us with a still limited dataset, generally related to regional studies on a restricted
98 scale of investigation.

99 In this paper, we discuss the first comprehensive data set of high precision $^{234}\text{U}/^{238}\text{U}$ ratios
100 and uranium concentrations measured by thermal ionisation mass spectrometry (TIMS), in
101 groundwater samples from the different aquifer levels of the Tunisian CT. The uranium data
102 are reported and analyzed along with a review of the isotopic composition of water ($\delta^2\text{H}$,
103 $\delta^{18}\text{O}$), and dissolved carbon (^{14}C and $\delta^{13}\text{C}$) measured in the same samples. Our two objectives
104 are first to improve our understanding of the mechanisms controlling the behavior and the

105 isotopic composition of uranium, and thus its potential use as a tracer in groundwater
106 hydrology and chemistry, and second to refine previous descriptions of the CT large-scale
107 circulation flowpaths and paleo-climatic evolution.

108 **2. Description of the study area**

109 The study area is located in southern Tunisia and covers the Chotts basin. This endoreic area
110 is limited to the East by the Tebaga-Kebili and Dahar mountains, to the west by the Algerian
111 border, to the north by the northern upland of the Chotts and Mélaoui upland, and in the
112 South by the Saharan platform (Fig.1).

113 **Fig.1**

114 The geology of the studied area has been investigated elsewhere (e.g. Castany., 1954;
115 Burollet., 1956; Busson., 1970; Zargouni et al., 1985; Bouaziz., 1995) and we will only recall
116 here the information relevant to the present work. Embedded under the Chott basin, the North
117 Western Aquifer System (NWAS) consists of two principal deep aquifers, the Continental
118 Intercalaire (CI) and the Complexe Terminal (CT), and of Quaternary shallow phreatic
119 aquifers. The CI is a multi layered aquifer composed of a succession of different sedimentary
120 series of late Jurassic to early Cretaceous detrital sediments, separated by gypsum
121 intercalation and clayey levels (Busson, 1967; Bouaziz, 1986). The groundwater reservoir of
122 the CI is contained in sandstone strata of Albo-Aptian and barremian deposits.

123 The formations which host the intermediate Complexe Terminal aquifer are Upper Cretaceous
124 carbonates and Paleogene/Neogene sands. These Upper Cretaceous strata, which are
125 dominated by mudstones, evaporites and carbonates, are overlain by Paleocene and Eocene
126 successions consisting of clays, calcareous shales with interbedded layers of gypsum,
127 dolomite and phosphatic limestones (Zouaghi et al., 2005). In some areas, Paleocene and
128 Eocene deposits constitute the substratum of the Mio-Pliocene sandy strata which, together
129 with the Upper Cretaceous carbonates, form the principal formations of the intermediate

130 Complexe Terminal aquifer (Buroillet, 1956; Biely et al., 1972; Mannai-Tayeche, 2009).
131 Elsewhere in the east of the Tozeur region, Mio-Pliocene sequences overlie unconformably
132 the Upper Cretaceous strata due to the erosion of the Paleocene and Eocene deposits
133 (Chaabani, 1995; Kamel, 2007). The Miocene series are overlain by Pliocene to Quaternary
134 continental deposits known as the Segui Fm that consists of a gypseous silty to sandy
135 succession becoming conglomeratic at the top and constitutes the upper shallow aquifer
136 (Castany, 1954; Buroillet, 1956; Zargouni et al., 1985; Tlig et al., 1991).

137 Conceptual block diagrams showing the different hydrostratigraphic units have been
138 elaborated to define the layering of the different aquifers and their connectivity, by
139 considering lateral facies changes and local tectonic structures (Fig.2.a and b).

140 The CT aquifer system is characterized by large changes in facies and thickness from North to
141 South and from East to West. The CT multi-layer aquifer is present both under the Nefzaoua
142 basin where it is hosted in Upper Senonian and Turonian carbonate formations (Fig.2.a) and
143 under the Djerid basin where Mio-Pliocene sands of Redjime Mâatoug, Tozeur and Chott
144 Gharsa plain constitutes the main aquifer level (Fig.2.b).

145 The Senonian and Turonian formations become deeper in the Djerid basin. The Senonian ones
146 are directly overlain by the Mio-Pliocene sands in the East of Tozeur region, while further
147 West, they are separated by phosphatic Eocene limestone strata (ERESS, 1972; Zargouni et
148 al., 1985). The Turonian consists of fractured and karstified dolomite/limestone with a mean
149 thickness of about 50 m. Groundwater flows mostly from Southeast to Northwest, broadly
150 from the Dahar mountains which constitute the main recharge area, to the main discharge area
151 in the Chott Djerid depression (Fig.1) (ERESS, 1972; OSS, 2003; Zammouri et al., 2007,
152 Hadj Ammar et al., 2014).

153 In the Djerid basin, Mio-Pliocene formations outcrop at the Tozeur uplift and become deeper
154 to the West of the basin. In this basin, groundwater flows from the southwestern Algerian
155 Great Oriental Erg, from the northern upland of the Chotts and from the Metlaoui upland.
156 Discharge areas are located in both the Chott Djerid and Chott Gharsa depressions (Fig.1)
157 (Edmunds et al., 1997; Kamel et al., 2007; Hadj Ammar et al., 2014).

158 The CT aquifer is confined over the majority of the Nefzaoua and Djerid basin. In fact, the
159 geological formations of the CT aquifer are overlain by clayey impermeable and/or sandy-
160 clayey semi-impermeable layers that restrict the vertical movement of water and thus, mixing
161 with water from the shallow aquifer is limited. However, at the proximity of faults,
162 preferential vertical leakage can occur between sandstones and carbonate layers of the CT
163 system as well as between the deep CI or the shallow aquifers and the CT system (Fig.2.a and
164 b). Only in the recharge area of Douz-Jemna regions of Nefzaoua basin and Kriz-El Hamma-
165 Degache areas of Djerid basin, the CT aquifer is under non-confined conditions. A possible
166 exchange between the CT and the shallow aquifer can thus occur in these locations (ERESS,
167 1972; Mamou, 1990; Kamel et al., 2007; Zammouri et al., 2007).

168 **Fig.2**

169 Historically, most of CT wells were artesian. Over the last two decades, an increasing
170 withdrawal for agriculture, industry and domestic uses have led to the overexploitation of
171 groundwater resources and caused a general drop of the water table (OSS, 2003). Nowadays,
172 only a few wells are still artesian.

173 **3. Sampling and analytical method**

174 The main hydrogeological units of the study area have been sampled during the spring season.
175 Three boreholes (#1, #2 and #3) from the Turonian aquifer level, twenty one samples from the
176 Senonian limestone aquifer (#4, #5,..., #24), seven boreholes from the Mio-Pliocene aquifer

177 of Redjime Mâatoug (#25, to #31), twenty four boreholes from the sandy Mio-Pliocene
178 aquifer of Tozeur (#32, to #55) and eleven boreholes from sandy Mio-Pliocene aquifer of
179 Chott El Gharsa plain (#56, #57, ..., #66) were sampled. The sampling grid has been chosen
180 as representative of the entire basin (Fig.1). We paid special attention to sample the different
181 lithological formations, at different depths, and cover the recharge and discharge zones of the
182 multi-layer aquifer.

183 All samples were collected directly from the well-head of currently exploited boreholes.
184 Groundwater was pumped until stabilization of temperature, pH and EC before sampling.
185 Screened interval depths mentioned in Table 1 correspond to the top of the screen.

186 Groundwater samples were filtered through 0.45 μ m acetate cellulose filter. Temperature,
187 electrical conductivity and pH were measured on site using a HI 9828 Multi-parameter probe.
188 Samples for the determination of the major element concentrations, as well as uranium
189 isotopic composition were stored in polyethylene bottles previously cleaned with diluted nitric
190 acid and distilled water and rinsed three times with sampled groundwater prior to filling.
191 Nitric acid was added to aliquots devoted to cations analyses. Volumetric titrations were
192 performed for the determination of bicarbonate alkalinity. Samples for radiocarbon analyses
193 were collected in the field by precipitating dissolved inorganic carbon as BaCO₃ from 100-
194 150 L sample volume, depending upon the alkalinity of each sample. The precipitates were
195 preserved in polyethylene bottles.

196 Major element concentrations, as well as water stable isotopes and ¹⁴C activities, were
197 measured at the Laboratory of Radio-Analysis and Environment of the National School of
198 Engineering of Sfax (Tunisia). Cations were analyzed with a ion chromatography equipped
199 with columns IC-Pak™ CM/D for cations, using EDTA and nitric acid as eluent, and anions
200 with a Metrohm chromatograph equipped with columns CI SUPER-SEP using phthalic acid

201 and acetonitrilic as eluent. The overall detection limit for each major element was about 0.04
202 mg/l. For each analysis, the ionic balance was lower than 5% (Table 1).

203 **Table 1**

204 Analyses of stable isotope ratios (^2H and ^{18}O) were performed by Laser Spectrometry with a
205 LGR DLT100 instrument (Penna et al., 2010) and reported in ‰ vs VSMOW standard
206 (Vienna-Standard Mean Oceanic Water) with a precision of $\pm 0.1\text{‰}$ for ^{18}O and $\pm 1\text{‰}$ for ^2H
207 (Table 2).

208 Radiocarbon activities were determined by benzene synthesis and liquid scintillation
209 spectrometry. Radiocarbon activities are reported as percent of modern carbon (pMC)
210 (Stuiver and Polach, 1977) with uncertainties of ± 1 pMC. Aggarwal et al. (2013)
211 demonstrated that uptake of atmospheric CO_2 may occur during the on-site precipitation
212 procedure on large volume alkaline samples, resulting in contamination by modern ^{14}C
213 potentially as high as 1 to 10 pMC. This represents an important caveat on our data that must
214 therefore be discussed with caution.

215 The $^{13}\text{C}/^{12}\text{C}$ ratios were determined by isotope ratio mass spectrometry at the IAEA and
216 CEREGE laboratories. Results are reported as δ values relative to VPDB (Vienna Pee Dee
217 Belemnite) standard. The ^{13}C uncertainty is of $\pm 0.3\text{‰}$ (Table 2).

218 Uranium isotopic compositions were determined by Thermal Ionization Mass Spectrometry
219 (TIMS; VG54-30) at CEREGE. For each sample, 200 ml were weighed and spiked with a
220 $^{233}\text{U}/^{236}\text{U}$ spike. After preconcentration by coprecipitation of iron oxy-hydroxyde, uranium
221 was separated and purified using UTEVA resin (Uranium and TEtraValents Actinides)
222 conditioned with nitric acid. The solution was evaporated, and the sample loaded on a single
223 rhenium filament with colloidal graphite. Masses 233, 234, 235 and 236 were measured in
224 peak-jumping mode on a VG54-30 mass spectrometer equipped with an ion-counting Daly
225 detector and a 30 cm electrostatic analyser. The activity ratio ($^{234}\text{U}/^{238}\text{U}$) was calculated by

226 assuming the natural value of 137.88 for the $^{238}\text{U}/^{235}\text{U}$ atomic ratio (Condon et al. 2010). The
227 uncertainties of the $^{234}\text{U}/^{238}\text{U}$ ratios were generally about 5‰ except for a few samples, for
228 which it could reach 9‰. The reproducibility of the U isotopic analyses was assessed with
229 replicate analyses of one sample ("Jemna" sample), which all agreed within error, and yielded
230 a mean value of $(^{234}\text{U}/^{238}\text{U}) = 1.668 \pm 0.004$ (2σ , $n = 3$). Total blanks for uranium were better
231 than 20 pg, thus negligible compared to the U amounts analysed in the sample (Table 2).

232 **Table 2**

233 **4. Results**

234 Hydrochemical, stable isotopes content and ^{14}C data, together with the uranium
235 concentrations and the $(^{234}\text{U}/^{238}\text{U})$ activity ratios are reported in Table 1 and Table 2.

236 **4.1. Geochemical characteristics of CT groundwater**

237 The major processes controlling the CT groundwater chemistry have been investigated in
238 previous studies (e.g., Abid et al., 2011, Jarraya et al., 2013, Hadj Ammar et al., 2014; Hadj
239 Ammar, 2016). In this paper, only specific features that could affect uranium geochemistry
240 will be highlighted. TDS values are ranging from 648 to 6290 mg/l and the major ions are in
241 the following order of contribution: calcium, sodium, chloride, sulfate, magnesium, potassium
242 and bicarbonate. Groundwater is mostly of the $\text{Na}^+\text{-Cl}(\text{SO}_4^{2-})$ and $\text{Ca}^{2+}(\text{Mg}^{2+})\text{-Cl}(\text{SO}_4^{2-})$
243 types. Groundwater geochemistry is controlled by water/rock interactions, mineral dissolution
244 and ion exchange (Hadj Ammar et al., 2014). The majority of water samples are dominated by
245 the dissolution of gypsum and halite, revealed by correlations between $[\text{Ca}^{2+}]$ and $[\text{SO}_4^{2-}]$ or
246 between $[\text{Na}^+]$ and $[\text{Cl}^-]$ among various sets of samples, and are saturated with respect to
247 dolomite and calcite precipitation (Table 1). High concentrations of the major ions observed
248 in some localities are explained by vertical mixing between surface and/or deep natural saline
249 water, induced by anthropogenic activities (Hadj Ammar et al., 2014).

250

251 **4.2.Dissolved uranium and $^{234}\text{U}/^{238}\text{U}$ ratio in CT groundwaters**

252 The results are illustrated in Figure 3, as $^{234}\text{U}/^{238}\text{U}$ activity ratios plotted versus $1/[\text{U}]$ (ppb)⁻¹.

253 Some of the main U-results are indicated along two representative cross-sections along a
254 generalized flow path in both Nefzaoua and Djerid basin (Fig.4a, b).

255 Uranium concentrations show a large variability from 1.5 to 19 ppb.

256 **Fig.3**

257 Most samples are in fact below 8 ppb, except four samples from the Mio-Pliocene sand
258 aquifer (#53 and #54 in Tozeur basin, and #57 and #62 in the Chott El Gharsa plain). The
259 lowest concentrations are found in two samples from the Senonian (#14 and #18) closest to
260 the aquifer outcrop in the Dahar mountain (Fig.4a), where active or recent recharge has been
261 identified by previous studies (Edmunds et al., 2003, Guendouz et al., 2003; Abid et al., 2010;
262 Abid et al., 2011). By contrast, the Turonian aquifer sample which is also located in this
263 recharge area, furthest to the East in the Dahar mountain, is among the highest uranium
264 content (#1, 8 ppb). These concentrations are in the upper half of the global distribution of
265 dissolved uranium in continental waters (Ivanovitch and Harmon, 1992), although without
266 extremely high values as high as 10-100 ppm sometimes encountered in uraniferous
267 environments. Such intermediate range is generally considered as associated with oxic or
268 mildly reducing conditions (Méjean et al., 2016; Riedel and Kübeck, 2018). By comparison
269 we measured much lower concentrations in the CI aquifer, well below 1 ppb and down to our
270 blank level, in the anoxic deepest regions of the basin (Chkir and zouari, 2007; Chkir et al.,
271 2009, 2012; Pertersen et al., 2013).

272 The $^{234}\text{U}/^{238}\text{U}$ activity ratios are all greater than one, ranging from 1.1 to 3.2. Thus, it appears
273 that all the samples, including those from limestone levels, are in radioactive disequilibrium
274 with excess of ^{234}U . A clear-cut distinction is observed between the Turonian and Senonian

275 carbonates aquifer, on the one hand, with $^{234}\text{U}/^{238}\text{U}$ activity ratios between 1.1 and 1.8, and
276 the Mio-Pliocene aquifer from Tozeur on the other hand, with higher ratios from 1.8 to 3.2.
277 Also noticeable is the striking contrast between the two groups of samples from the Mio-
278 Pliocene aquifer, as the samples from Chott El Gharsa plain, by contrast with those from
279 Tozeur basin, all cluster at the low $^{234}\text{U}/^{238}\text{U}$ and high [U] end-member in the diagram of
280 Figure 3, with the only exception of sample #58 at $^{234}\text{U}/^{238}\text{U} = 2.8$.

281 The highest $^{234}\text{U}/^{238}\text{U}$ activity ratios correspond to samples #36 and #52 of the Mio-Pliocene
282 sand close to Tozeur uplift, and also to sample #25 of Mio-Pliocene in the western part of the
283 Redjime Mâatoug region (Fig.4b).

284 **Fig.4**

285 **4.3.Stable isotopes and groundwater ages**

286 The stable isotope data of the same samples as in this study have been reported previously by
287 Hadj Ammar et al., 2014. $\delta^{18}\text{O}$ ranged from -7.8‰ to -4.2 and $\delta^2\text{H}$ from -57.4‰ to -34.8‰,
288 with mean values of $(-6.0 \pm 0.2\text{‰})$ and $(-46.1 \pm 1\text{‰})$ respectively (Table 2). This isotopic
289 variation in CT groundwaters is controlled dominantly by evaporation processes, paleo-
290 recharge and mixing between waters with different isotopic ratios (Fig.12 in Hadj Ammar et
291 al., 2014).

292 Radiocarbon activities and $\delta^{13}\text{C}$ values of DIC determined for 34 water samples cover a wide
293 range from 30.2 to 1.1 pmc and from -10.75 to -3.6‰, respectively (Table 2, Fig.5) (this
294 work, and Abid et al., 2011; Abid et al., 2012; Yangui et al., 2012). The relatively high $\delta^{13}\text{C}$
295 values shows that dissolution of solid carbonate and carbon exchange between DIC and
296 carbonate minerals are the dominant processes affecting the $\delta^{13}\text{C}$ and ^{14}C content of DIC in
297 the CT groundwater.

298 **Fig.5**

299 A large number of correction models, relying on various assumptions, have been proposed in
300 the literature for dating groundwater with radiocarbon, in order to account for the different
301 processes which may affect the ^{14}C activity through dissolution/precipitation processes with
302 the basement lithology, or exchange with the gas phase in the unsaturated zone (Ingerson and
303 Pearson, 1964; Evans et al., 1979; Fontes and Garnier, 1979; Salem et al., 1980; Eichinger,
304 1983; Clark and Fritz, 1997, Buckau et al., 2000a; Zhu and Murphy, 2000; Geyh, 2000, 2005;
305 Han and Plummer, 2013, 2016).

306 The models cited above have been widely applied and discussed in the literature and are
307 therefore not discussed in detail here. In this study, we used the model of Fontes and Garnier
308 (1979) which takes into account the dissolution and precipitation of carbonates and
309 evaporites, the Ca/Na ion exchange process on clay minerals and the transition from open to
310 closed conditions for carbonate dissolution. A detailed explanation on the choice of this
311 model among the others and the following C signatures used to calculate radiocarbon ages of
312 southern Tunisia groundwaters were well discussed in Abid et al., 2012, 2014. The model
313 ages that we obtain are listed in Table 2 and illustrated as a frequency histogram based on
314 Gaussian distribution analysis within a ($\pm 3\sigma$) interval (Fig.6). Although widely scattered
315 between 1.5 and 25 ka, the distribution of model ages seems to show two main broad clusters
316 centered around 5 ka and 13 ka respectively. The youngest ages (1.5 – 2.5 ka) are found in
317 Senonian and one Turonian samples, and the oldest (19 – 25 ka) are all from the Tozeur basin
318 Miocene aquifer.

319 **Fig.6**

320 **5. Discussion**

321 Each of the five regional aquifers studied here show a distinct pattern in the $^{234}\text{U}/^{238}\text{U}$ versus
322 $1/[U]$ diagram of Figure 3.

323 **5.1. Southern aquifers: Nefzaoua and Redjime Mâatoug**

324 For the two aquifers from the southern part of the basin, i.e. Senonian from Nefzaoua basin
325 and Miocene from Redjime Mâatoug, a clear geographical repartition is noted in the data
326 (Fig.4, Fig.7). In the first case, U-concentration increases along the groundwater pathway
327 inferred from the piezometry, as the aquifer deepens away from the recharge area in the Dahar
328 Mountain (#14 and #18), down to sample #15 and #17 to the north, #16 to the north-east, #12
329 to the south-west. Similarly, the lowest concentrations are found in the eastern part of
330 Redjime Mâatoug (#31 and #29), under the southern edge of Chott El Djerid, whereas higher
331 values are found further west, although in this second case with no clear relationship to
332 present-day piezometry.

333 **Fig.7**

334 Similar increases have been interpreted classically in various previous studies around the
335 world (e.g. Arendt et al., 1980; Spalding and Druliner, 1981) as resulting from progressive
336 dissolution of the basement rock along the transit of the groundwater. This explanation is
337 consistent with our data and is coherent with the observation that uranium is well correlated
338 with the total dissolved ions, and particularly with sulfate ($0,31 < R^2 < 0,91$) and calcium
339 ($0,69 < R^2 < 0,75$) (Fig. 8). Dissolution of gypsum and anhydrite have been suggested as a
340 dominant control of these groundwaters chemistries, especially in the case of Redjime
341 Mâatoug where $[Ca^{2+}]$ and $[SO_4^{2-}]$ are strongly correlated (Hadj Ammar et al., 2014).

342 **Fig.8**

343 The hypothesis that uranium could be associated with sulfate bearing evaporites must thus be
344 considered and would deserve further testing on the solid phase. Nevertheless, it must also be
345 noted that Nefzaoua groundwaters show systematically higher $[U]/[Ca^{2+}]$ and $[U]/[SO_4^{2-}]$
346 ratios than Redjime Maatoug in Fig.8, suggesting that more than one mineral phase is

347 releasing uranium to the solution. Indeed, for the Senonian groundwaters, the calcareous
348 lithology of the basement is expected to make a significant contribution to the dissolved
349 uranium content (e.g. Bonotto and Andrews, 2000). Other mechanisms are often advocated as
350 possible control factors of the water chemistry, such as dedolomitization or cation exchange
351 on bedrock surface (Hiscock, 2005; Chae et al., 2006; Escorcia et al., 2013; Luo et al., 2018).
352 However, neither of these two processes is apparent in our data, as suggested by the absence
353 of correlation between [U] and the Mg/Ca ratio, nor with the Na/Ca ratio (not shown).

354 Moreover, in both aquifers, the geographical trend is far from regular, with the highest
355 concentrations observed in intermediate positions along the transects, which suggests
356 heterogeneous and patchy distribution of the basement lithology, and also likely irregular
357 pathways and variable flow rates of the groundwater circulation.

358 By contrast with their comparable features for uranium concentrations, the two southern
359 aquifers show very contrasted trends in isotopic composition. The $^{234}\text{U}/^{238}\text{U}$ ratio increases
360 regularly in the Redjime Mâatoug samples along the concentration gradient, up to one of the
361 highest values in our data set (# 25: $^{234}\text{U}/^{238}\text{U} = 3.1$) (Fig.4b), while the Senonian samples do
362 not show such evolution with values randomly scattered within a narrow band between 1.5
363 and 1.8, except for sample #18 close to equilibrium ($^{234}\text{U}/^{238}\text{U} = 1.1$) (Fig.4a). The ^{234}U
364 excess is typical of all surface and underground continental waters and is a fairly well-known
365 phenomenon since the early compilations of Ivanovitch and Harmon (1992). It has been
366 documented in numerous environments, although comparatively less often in limestone
367 aquifers (Osmond and Cowart, 1976; Andrews and Kay, 1983; Osmond and Ivanovich, 1992;
368 Gascoyne, 1992; Davis and Krogh, 2001; Paces et al., 2002; Porcelli and Swarzenki, 2003;
369 Chabaux et al., 2003, 2008; Smedley et al., 2006). Our data from the Senonian limestone
370 aquifer are thus interesting in two main aspects: first they confirm the isotopic disequilibrium
371 of uranium dissolved from calcareous formations, showing that α -recoil processes are also

372 significant in such facies even though water-rock interaction mainly occurs as congruent
373 dissolution. Second, we do not observe the decreasing trend of $^{234}\text{U}/^{238}\text{U}$ with increasing
374 uranium content which was described in several studies (Kronfeld and Adams, 1974; Osmond
375 and Coward, 1976; Burkitbayev et al., 2012). This inverse trend is generally interpreted as
376 reflecting progressive dampening of the ^{234}U fractionation due to alpha recoil by the increase
377 in uranium at secular equilibrium released as weathering increases. Instead, this constant
378 $^{234}\text{U}/^{238}\text{U}$ ratio suggests that the ratio between the rate of removal of ^{234}U from the rim of rock
379 affected by alpha recoil, and the rate of bulk chemical dissolution remains constant along the
380 groundwater pathway, as expected in a homogeneous basement (Fig.4a).

381 By contrast, the interpretation is more complex for the Mio-Pliocene aquifer from Redjime
382 Mâatoug, where the ^{234}U preferential leaching increases with [U] (Fig.4b) . A possible simple
383 explanation could advocate mixing in various proportions between two groundwater end-
384 members with contrasting composition, related to the type of dominant basement rock
385 encountered along their pathway, e.g. carbonates for the (low [U]-low $^{234}\text{U}/^{238}\text{U}$) end-
386 member, and evaporite (mostly of sulfate-bearing type) for the (high [U]-high $^{234}\text{U}/^{238}\text{U}$) end-
387 member. Under this assumption, the regional trend observed in the data would reflect a
388 progressive increase of the proportion of evaporites within the aquifer reservoir along the
389 East-West gradient of the Redjime Maâtoug basin.

390 ***5.2. Northern sectors: Chott El Gharsa plain and Tozeur ridge***

391 Compared to the two southern aquifers, the two northern Mio-Pliocene sandy aquifers show
392 more complex patterns, and quite different from each other.

393 On the one hand, the seven samples from the Mio-Pliocene of the Chott El Gharsa plain all
394 plot in a relatively restricted domain in Figure 3, corresponding to the (high [U], low
395 $^{234}\text{U}/^{238}\text{U}$) end-member, similar to the most enriched Senonian and Turonian samples: two of
396 these samples (#57 and #62), along with one of Turonian age, bear indeed the lowest

397 $^{234}\text{U}/^{238}\text{U}$ in our study. The similarity between the sandy aquifer of Chott El Gharsa plain and
398 the calcareous layers further South is unexpected. This seems to suggest counter-intuitively
399 similar U-bearing phases and similar duration of water-rock interaction in both cases. The
400 flow lines inferred from the present piezometry indicate a circulation oriented to the South in
401 this region, and thus a recharge further North, possibly around the Metlaoui upland. The
402 transit distance of the groundwater is thus probably comparable with that of the southern
403 aquifers, and the depth of collection significantly deeper (500-1100 m depth), thus possibly
404 coherent with longer residence times and higher dissolved mineral load.

405 Finally, the 24 samples collected from the sandy aquifer along the Tozeur uplift and the
406 Tozeur basin show the widest dispersion in Figure 3, encompassing both the highest results in
407 $^{234}\text{U}/^{238}\text{U}$ ratios (#36 and 52) and highest [U] (#53 and 54), but also samples trending towards
408 the depleted end-member (#33, 35, 38 and 48), or towards the (high [U], low $^{234}\text{U}/^{238}\text{U}$) end-
409 member (#40 and 45), although all samples but one have $^{234}\text{U}/^{238}\text{U}$ ratios higher than 2. Such
410 a large diversity must be related to the complex geomorphology and tectonic structure of the
411 Tozeur uplift that is bordered or crossed by several large faults and which represents the
412 surface expression of a large uplift of the sedimentary strata at depth (Fig. 2.b). Moreover, the
413 Tozeur region is also known for a complex hydrogeological context, resulting in upwelling of
414 deep-seated groundwater attested by the occurrence of numerous springs and artesian wells
415 before the recent exponential increase of the extraction for human activities. Although widely
416 scattered in terms of spatial repartition, the results show nevertheless some significant
417 groupings. For instance, the four most depleted samples (#48, 38, 33 and 35) are all clustered
418 on the southern slope of the Tozeur uplift, in the north-eastern region (Fig.1, Fig.3 and
419 Fig.4a). These samples follow the same trend as that defined by those from Redjime Mâatoug
420 and seem to correspond to initial stages of evolution. This may mean either close proximity to
421 recharge sites, or slower recharge rates, or alternatively a basement lithology depleted in the

422 (high $^{234}\text{U}/^{238}\text{U}$, high [U]) source rock, supposedly associated with sulfates in the case of
423 Redjime Mâatoug.

424 Similarly, another series of samples located further West in the Tozeur plain (# 36, 52, 46, 37)
425 are regularly spaced along a linear trend between the (high $^{234}\text{U}/^{238}\text{U}$, high [U]) and (low
426 $^{234}\text{U}/^{238}\text{U}$, high [U]) end-members. This suggests that the two types of waters sustaining these
427 end-members, i.e. represented at the end of the Redjime Mâatoug transect on the one hand,
428 and under the Chott El Gharsa plain on the other, are also present under the Tozeur plain and
429 mix in various proportions either along strata or through inter-layers connections.

430 Finally, a last noticeable observation is that the two samples most enriched in uranium (#53
431 and 54), which lie clearly above the general correlation between [U] and TDS in Fig. 8, are
432 associated with the phosphate ore of Tozeur-Nefta located on the northern edge of the Tozeur
433 ridge (Fig. 1) and embedded in Eocene layers directly underlying the Miocene CT aquifer
434 (Gallala et al., 2016). These phosphates are rich in uranium (35 ppm, Sassi, 1974; Chaabani,
435 1995). Uranium is known to have a high affinity for phosphates, as well as for carbonates,
436 which can also exert a strong control on its behavior in solution (Sandino and Bruno, 1992;
437 Adam et al., 2011; Beazley et al., 2011; Schipper et al., 2011; Alloway, 2013; Mehta et al.,
438 2014; Cumberland et al., 2016). As for the previous examples, the contact between the
439 groundwater and the source rock must be very localized, along fractures or channels, since
440 sample #50, also very close to the same site, do not show the same enrichment. Again,
441 experimental leaching experiments on solid phases, as well as phosphorus measurements in
442 the waters would be necessary before a definite interpretation of these results.

443 ***5.3. Uranium concentrations and isotopes versus major element chemistry***

444 In addition, it is worth underlining that the different trends discussed above, based on uranium
445 content and isotopic ratio, do not correspond to equivalent grouping in major element
446 composition. The chemical results are indeed randomly scattered between the 1) dilute

447 recharge waters, 2) sulfate end-member and 3) chloride end-member, reflecting the main
448 control of evaporites dissolution on the groundwater chemistry (Hadj Ammar et al., 2014).
449 The four regional aquifers discussed above show large overlaps in their chemical signatures,
450 and the sulfate and chloride types are both represented in each of them. The Chott El Gharsa
451 samples are all in the upper range of total dissolved ions, the Tozeur uplift samples are highly
452 dispersed, and the most dilute samples are found in the Senonian Nefzaoua samples and
453 Miocene Redjime Mâatoug in the south-eastern region. There is no general correlation
454 between the uranium concentration and any of the major ions, except for the qualitative
455 observation already noted above that the highest [U] also correspond to the highest sulfate
456 contents (# 53, 54, Fig. 8). By comparison, samples most enriched in Cl, like Mio-Pliocene
457 #39 and #43, or in Ca like # 36 and #39, are not particularly rich in dissolved uranium (Fig.8).
458 Finally, a remarkable feature of these data is that the high $^{234}\text{U}/^{238}\text{U}$ end-member samples,
459 such as Mio-Pliocene #36 and #52 of the Tozeur basin, or Mio-Pliocene #25 of the Redjime
460 Mâatoug region, do not bear any particular chemical signature in their major ion composition
461 (Fig. 4b, Fig. 8). High $^{234}\text{U}/^{238}\text{U}$ ratios have often been interpreted as resulting from a
462 downstream position of the sample with respect to a redox-front across the aquifer (Osmond
463 and Cowart, 2000; Petersen et al., 2013; Méjean et al., 2016), where the alpha-recoiled ^{234}U
464 continues to be leached out of the basement rock while uranium has been quantitatively
465 depleted by precipitation in the reductive environment. However, this interpretation seems to
466 be ruled out in the case of the CT groundwaters, characterized generally by medium or high
467 uranium contents typical of oxic conditions. A possible clue lies in the observation that all the
468 samples with large ^{234}U excess are located in upstream positions with respect to present-day
469 piezometric lines (see Fig.1), as already noted for the Tozeur basin data, suggesting that the
470 radioactive disequilibrium might be linked to the early phase of water-rock interaction within
471 sandstone aquifers, although this hypothesis requires obviously further verification.

472 5.4. *Chronological and paleoenvironmental inferences*

473 Paleoclimatic studies revealed the occurrence of several humid phases over the entire
474 Mediterranean basin during the last glacial-interglacial cycle, based on marine sediments
475 (Fontugne et al., 1994), foraminifera (Kallel et al., 2000; Essallami et al., 2007), lacustrine
476 records (Ariztegui et al., 2000; Causse et al., 2003), and stalagmites (Genty et al., 2003, 2006;
477 Braun et al., 2019). Groundwaters may also constitute to some extent archives of these
478 climate oscillations. For instance, Petersen et al. (2014) showed that the distribution of
479 cosmogenic ^{36}Cl along the South Saharan CI pathway is strongly controlled by the alternation
480 of dry and humid phases, and that the last interglacial period must have been particularly
481 humid. This finding is strongly supported by ^{14}C measurements which are below the
482 detection limit that indicate residence times greater than 35 ka.

483 Our $\delta^{18}\text{O}$ and $\delta^2\text{H}$ data in the southern Tunisian CT (Hadj Ammar et al., 2014) showed that
484 the aquifer was recharged with water more depleted in heavy isotopes than present-day
485 precipitation, as generally observed in deep groundwaters over the whole Northern Africa.
486 This observation is also coherent with the recharge temperatures lower by 2-3°C than present,
487 estimated from noble gas paleothermometry in CT groundwater from Algeria (Guendouz et
488 al., 1997; Stute and Schlosser, 1993).

489 Our data also demonstrated that the different regional aquifers of the CT suffered from
490 various degrees of evaporation, indicating different hydroclimatic conditions upon recharge.
491 Specifically, the Chott El Gharsa CT groundwaters showed more depleted isotope signatures
492 than the other regions, close to the global meteoric water line, a characteristic shared with
493 three Senonian and with the Turonian samples (Fig.9). This similarity was already noted with
494 the uranium results, these samples constituting together the (low $^{234}\text{U}/^{238}\text{U}$, high [U]) end-
495 member in Figure 3. This confirms the striking contrast between the groundwaters from the
496 different sandy Mio-Pliocene aquifers, and the possible synchronicity between the Nefzaoua

497 and Chott El Gharsa recharge. By contrast, all the other regions are scattered towards less
498 depleted values along a common evaporation trend.

499 **Fig.9**

500 Both the stable isotope compositions and the ^{14}C activities of our samples are thus consistent
501 with recharge periods scattered over the Holocene and Late Pleistocene. ^{14}C model ages,
502 although strongly model-dependent and prone to contamination bias, show a noticeable
503 geographical repartition with younger model ages located in the south-eastern Senonian
504 aquifer, thus compatible with the general circulation pattern inferred from present-day
505 piezometry, which suggests a SE-NW flow path originating from the Dahar. At the other end,
506 the oldest ^{14}C model ages are scattered in the deepest regions of the basin, on the northern
507 edge of Chott El Gharsa and under the Tozeur plain and Tozeur uplift (Fig.6 and Fig.10).

508 On the other hand, the ^{14}C activities measured in the samples from the Dahar upland is only
509 slightly lower than 30 pMC, which indicates either an early Holocene recharge, or mixing
510 between a small fraction of modern recharge with more ancient waters. The second
511 hypothesis seems to be confirmed by the presence of low but detectable environmental tritium
512 contents in samples #14 (0.7 TU) and #18 (0.45 TU) (Hadj Ammar, 2016). The low ^{14}C
513 activity was also observed in one sample collected from the nearby outcrop of the CI by
514 (Petersen et al. 2018). The high $\delta^{13}\text{C}$ values measured in all our samples, including those with
515 the youngest ^{14}C model ages, also demonstrates that the addition of dead carbon leached out
516 of the carbonates from the reservoir formation happens early during the groundwater
517 percolation along the aquifer layer.

518 **Fig.10**

519 As detailed above in the different regional discussions, we do not identified any general
520 correlation between the uranium trends and these broad scale characteristics of the aquifer. In

521 particular, our data do not seem to corroborate the conclusions from recent studies correlating
522 uranium concentrations and $^{234}\text{U}/^{238}\text{U}$ activity ratios with the ^{14}C activities and residence time
523 (Kronfeld et al., 1994; Malov, 2013; Malov et al., 2016), who found that groundwater with
524 lower residence time, have enriched $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values and lower $^{234}\text{U}/^{238}\text{U}$ ratios.
525 Therefore, the complexities described above in the uranium isotope variations preclude their
526 use as a chronometer in this case as in most previous studies around the world, although with
527 several exceptions in very specific situations (e.g. Chalov et al., 1964, 1970; Kronfeld and
528 Adams, 1974; Andrews and Kay, 1978; Andrews et al., 1982).

529 **6. Conclusion**

530 We have produced in this study the largest data set available so far of uranium concentrations
531 and $^{234}\text{U}/^{238}\text{U}$ isotopic ratios in the Complexe Terminal of the Sub Saharan Aquifer System,
532 with samples representative of the full variety of lithological reservoirs and hydrological
533 situations encountered in this large-scale groundwater aquifer. The results show a large range
534 of variation, both for Uranium concentration (factor 13 between the most dilute and most
535 enriched samples), and for $^{234}\text{U}/^{238}\text{U}$ values, with all samples out of secular equilibrium with
536 excess ^{234}U ranging from 1.1 to 3.2. Moreover, the different regional sub-systems are
537 characterized by different trends clearly distinct from each other, by contrast with the major
538 ions chemistry which shows large overlaps. As a whole, the results are scattered among three
539 end-members: 1) (low [U], low $^{234}\text{U}/^{238}\text{U}$) typical of the southeastern samples closest to the
540 Dahar recharge area; 2) (high [U], low $^{234}\text{U}/^{238}\text{U}$) found in the Senonian and Turonian mostly
541 carbonated aquifer layers, and in the Miocene sandstone from the Chott El Gharsa; and 3)
542 (high or intermediate [U], high $^{234}\text{U}/^{238}\text{U}$) identified in some samples from the Redjime
543 Mâatoug Miocene sandstone in the Southwest region, and in the Miocene from the Tozeur
544 area. The origin of those variations remains largely speculative, awaiting for a definite
545 identification of the specific uranium bearing solids in the basement rock related to the different

546 end-members. In this respect, future investigations should probably aim at revisiting
547 experimental leaching of solid phases from drill cores, when available, with a special attention
548 to sulfate and phosphate bearing phases. Of particular interest is the observation that all the
549 groundwater samples from the limestone-dominant layers of the Senonian and Turonian are in
550 radioactive disequilibrium, seemingly unexpected as carbonate weathering is generally
551 considered mostly as a congruent dissolution process. The $^{234}\text{U}/^{238}\text{U}$ ratios are not correlated
552 with the concentration in uranium in this part of the CT, in contrast with previous studies in
553 other regions of the world, suggesting in this case a constant ratio between the uranium
554 addition due to weathering, and ^{234}U enrichment from alpha recoil. The exact mechanism of
555 this enrichment remains thus to be elucidated, linked either to the carbonate itself, or minor
556 phases, or even surface exchange reactions. On the other hand, the high $^{234}\text{U}/^{238}\text{U}$ values,
557 higher than 1.8 are only found in the groundwaters from the Miocene sandstones, in a patchy
558 distribution probably related to the highly heterogeneous lithology of the basement rocks.

559 More generally, no simple correlation is found between the hydrological features of the CT
560 and the uranium signature of the groundwaters, except in the upstream regions of the path-
561 flow closest to the Dahar recharge area. While ^{14}C model ages are broadly consistent with a
562 SE-NW direction of the groundwater circulation, coherent with the dipping of the sedimentary
563 layers in the basin, alternatively high or low $^{234}\text{U}/^{238}\text{U}$ values can be found in the deepest parts
564 of the aquifer.

565 Based on radiocarbon model ages and isotopic signatures, we infer that recharge of the CT
566 aquifer occurred during different climatic conditions compared to the present day. This
567 paleorecharge extended from the end of the last glacial to the Holocene periods.

568 In spite of the number of questions remaining unanswered at this stage about its behaviour,
569 the results of this study confirms that uranium bears a wealth of original informations about

570 the groundwater chemical evolution, and thus should deserve a dedicated attention in future
571 research in chemical hydrology.

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930 **Figure captions**

931 **Fig.1** Simplified map of the study area, showing sample location and groundwater
932 piezometric level of the CT aquifers.

933 **Fig.2** Block diagrams illustrating the compartmentalization of the different layers of the
934 intermediate CT aquifer in both Nefzaoua (a) and Djerid (b) basins with geological groups
935 and clarifying the probable mixture with the other shallow and deeper fractured aquifers.

936 **Fig.3** The $^{234}\text{U}/^{238}\text{U}$ activity ratio (AR) and the reciprocal of the U- concentration (in ppb^{-1}) in
937 the CT groundwaters (numbers correspond to the sample identification – Table 2).

938 **Fig.4** Conceptual hydrochemical cross-sections along the generalized flow path in Nefzaoua
939 (a) and Djerid (b) basins, showing some of the main U and major ions-results.

940 **Fig.5** Plots of ^{14}C activities versus $\delta^{13}\text{C}$. The small symbols indicate the A_0 initial values
941 calculated for each point with the Fontes and Garnier (1979) model. γ This work, na: not available,
942 * Abid et al., 2011, ι Yangui et al., 2012, ϵ Abid et al., 2012, Θ Hadj Ammar et al., 2014.

943 **Fig. 6** Frequency histogram of ^{14}C model ages calculated from Fontes and Garnier (1979).

944 **Fig. 7** Spatial distribution of U- concentration.

945 **Fig. 8** Major ion contents Versus U- concentration. Θ Hadj Ammar et al., 2014, γ This work

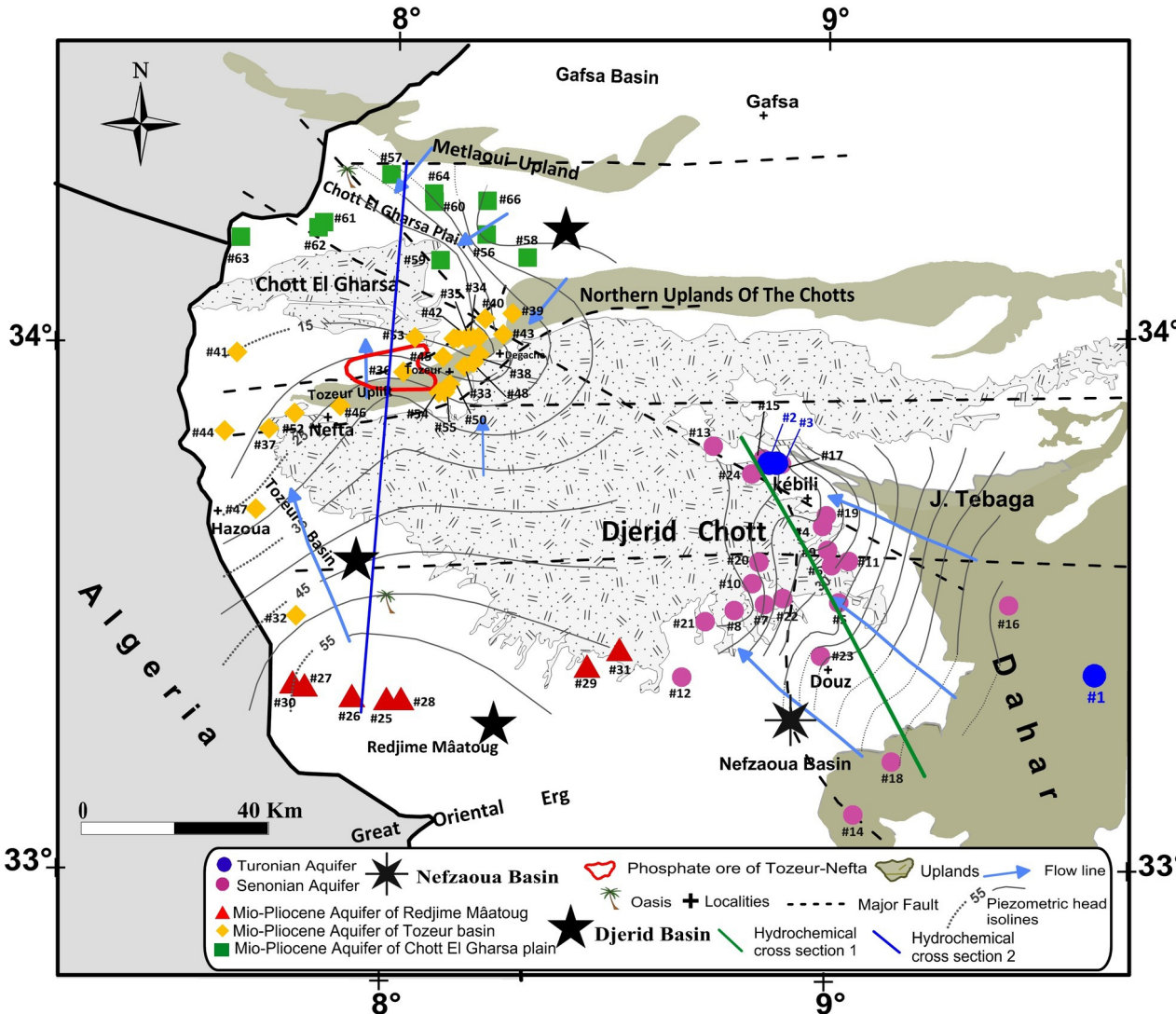
946 **Fig.9** Plots of $\delta^{18}\text{O}$ content versus U- concentration. Θ Hadj Ammar et al., 2014, γ This work

947 **Fig.10** Spatial distribution of the main recharge periods of the CT groundwater.

948 **Tables**

949 **Table 1.** Field parameters and chemical results of analyzed groundwater samples of the CT
950 aquifer in Chott Region.

951 **Table 2.** Uranium isotopic data together with stable isotopes and ^{14}C corrected age of
952 analyzed groundwater samples of the CT aquifer.



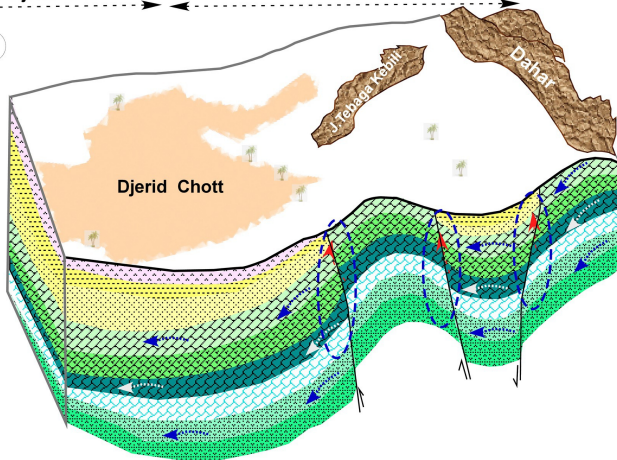
NW

SE

Djerid Basin

Nefzaoua Basin

a



Quaternary

Upper
Senonian

Cenomanian



Pliocene

Early
Senonian

Albo-Aptian



Miocene



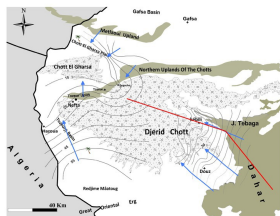
Turonian



Barremian

possible inter-layer
mixing zones

Groundwater flow

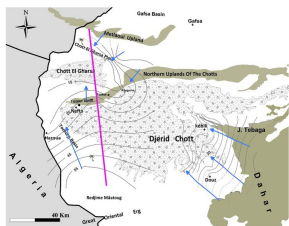
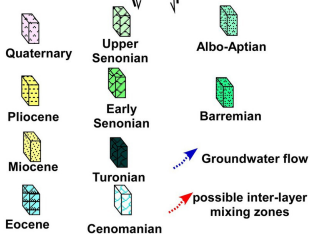
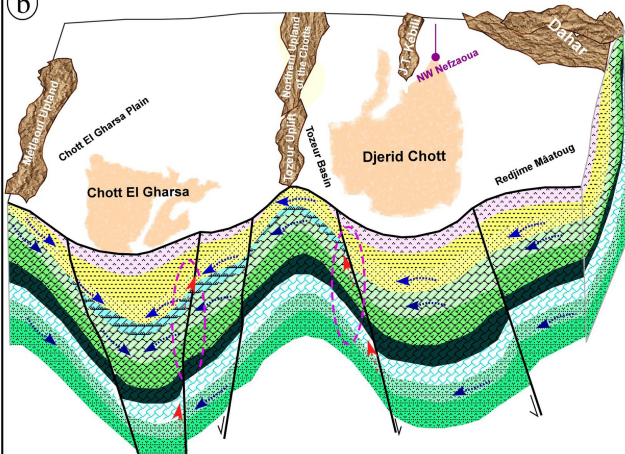


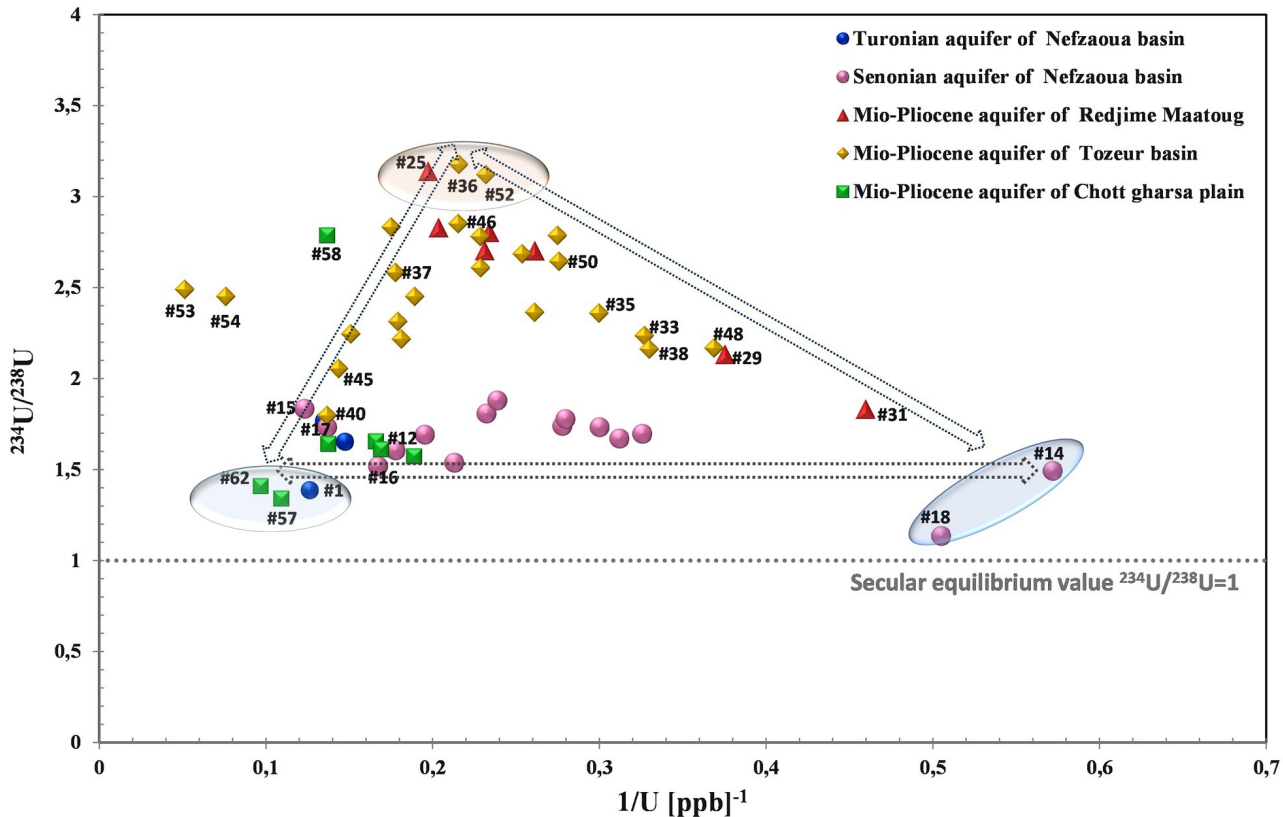
NW

Djerid Basin

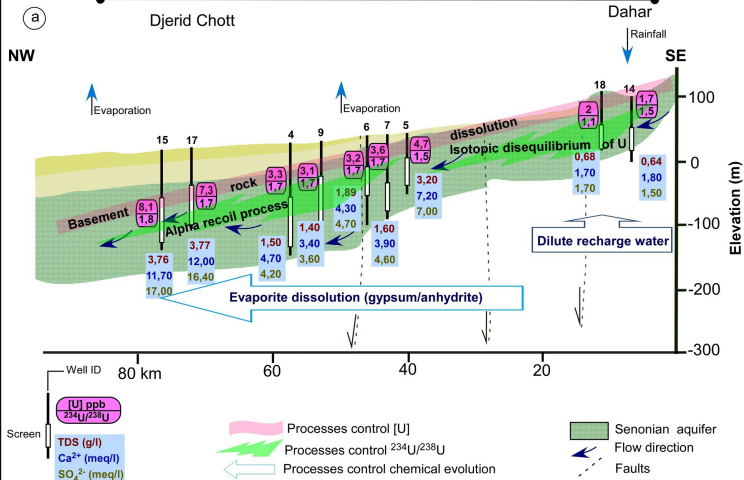
SE

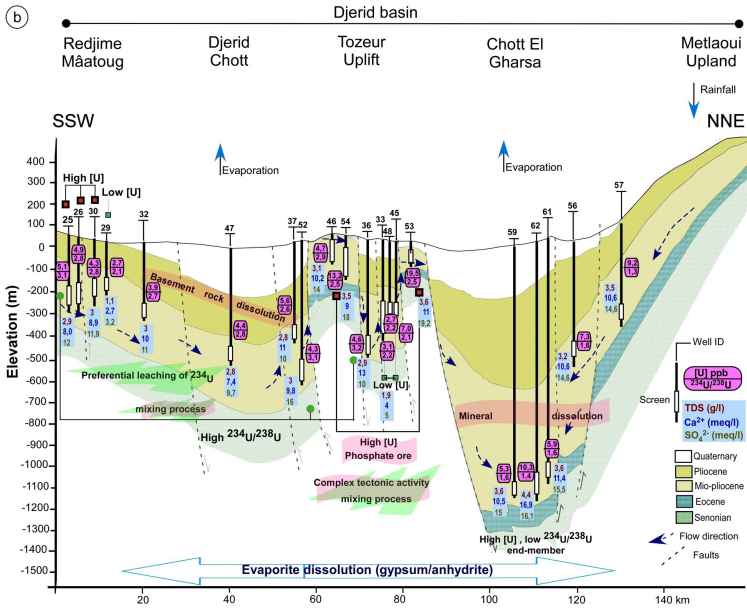
(b)

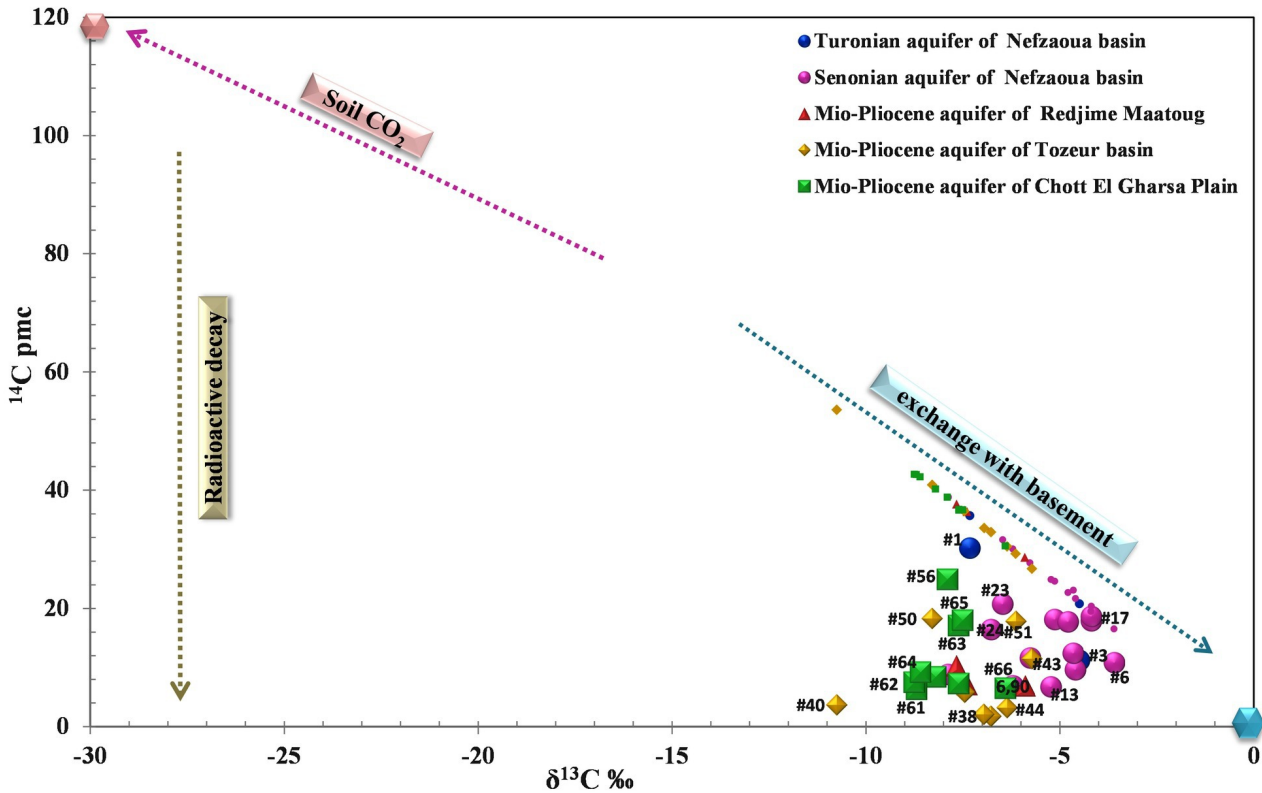


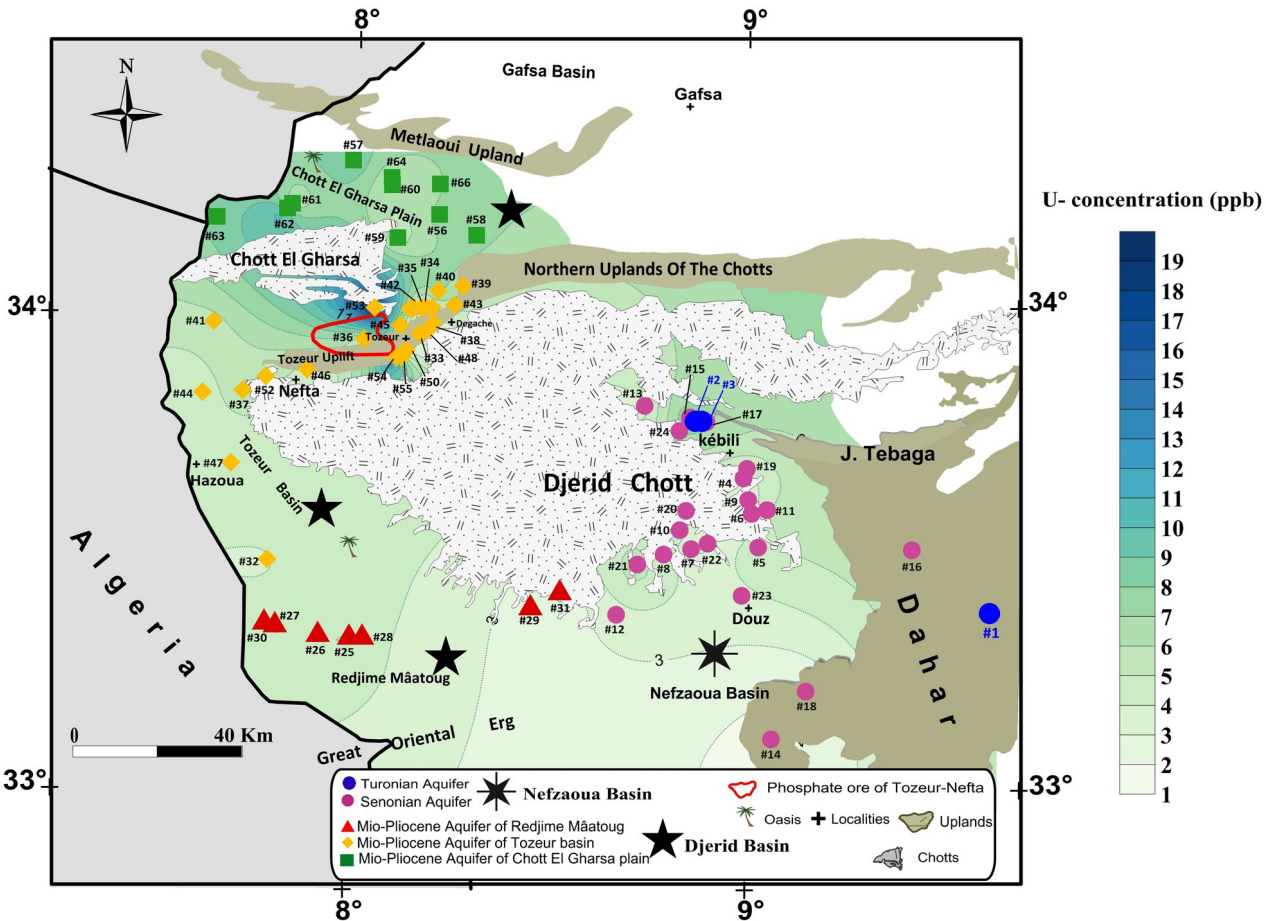


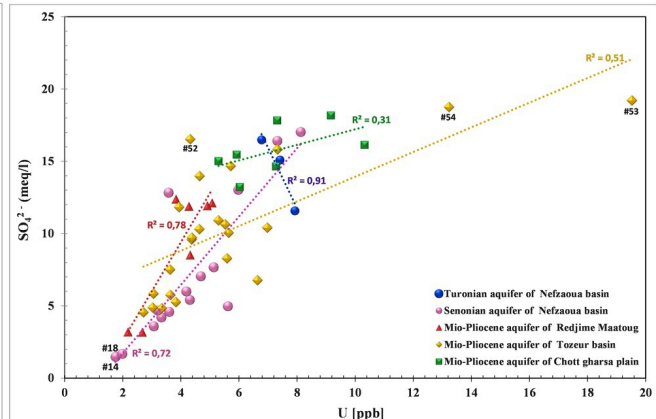
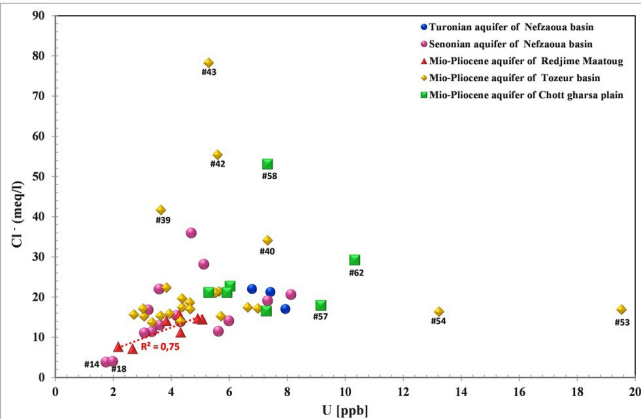
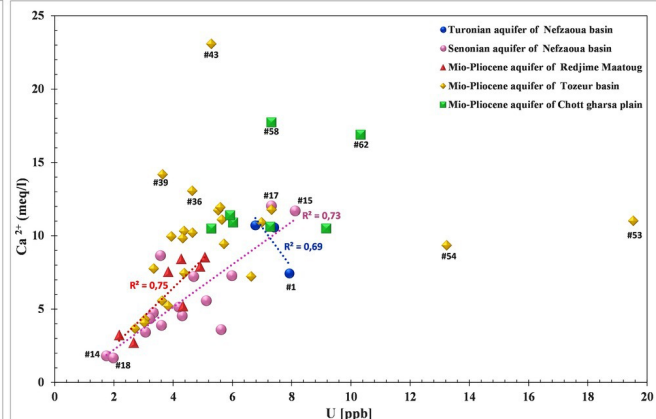
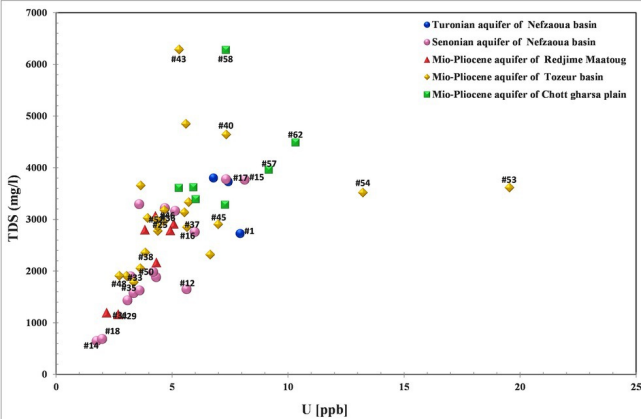
Nefzaoua basin

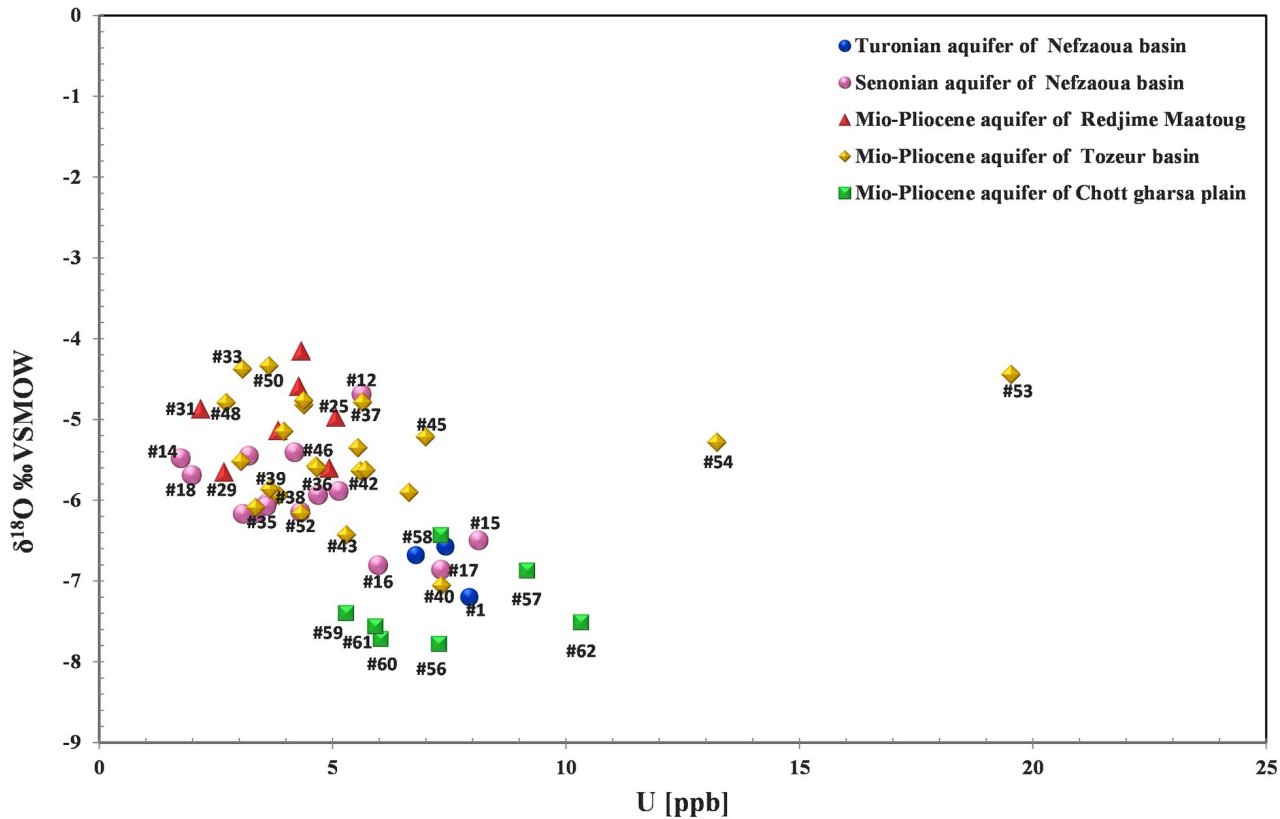












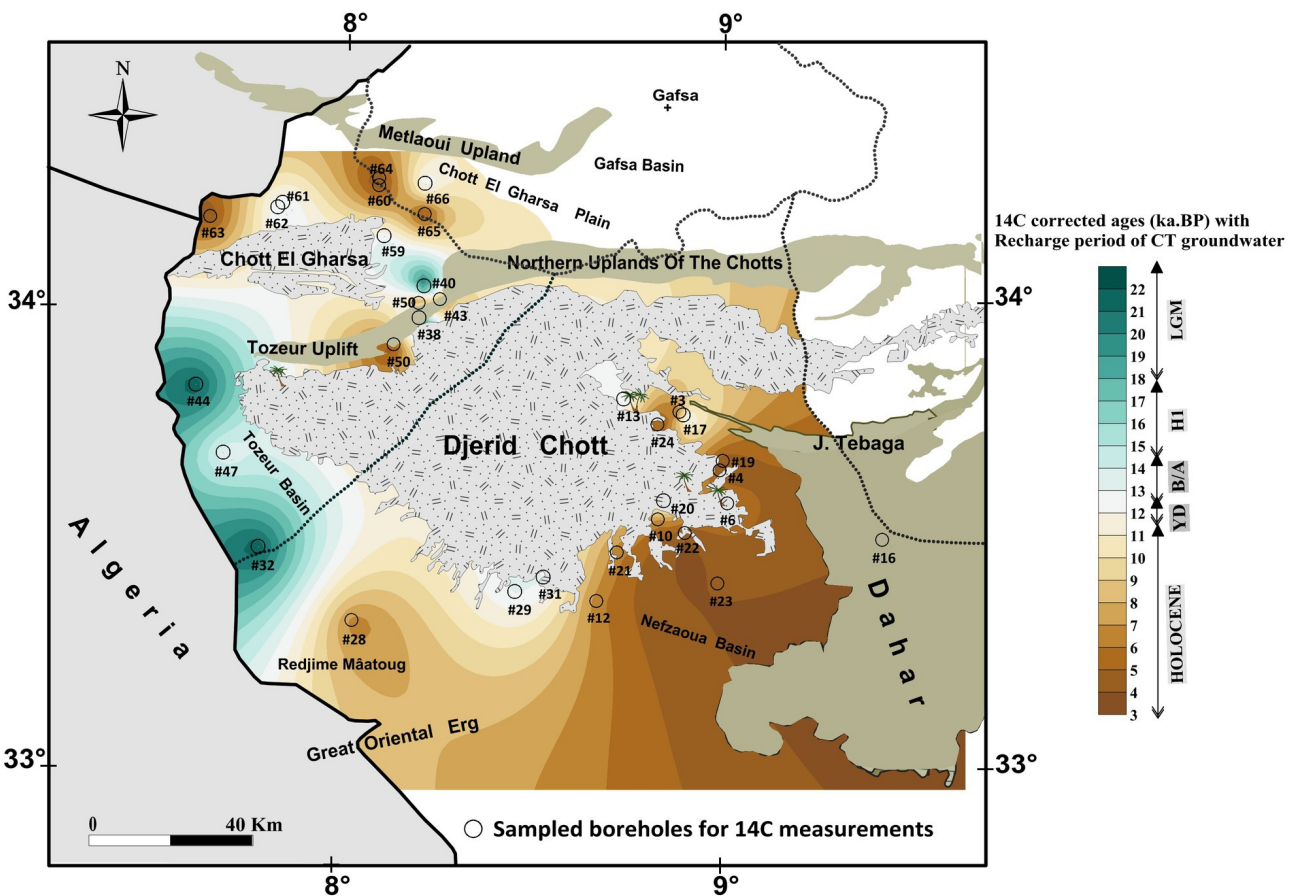


Table1. Field parameters and chemical results of analyzed groundwater samples of the CT aquifer in Chott Region (Θ).

Sample Labels	Aquifer	Basin	T ° C	pH	Screen depth (m)	Cl ⁻ (meq/l)	NO ₃ ⁻ (meq/l)	SO ₄ ²⁻ (meq/l)	HCO ₃ ⁻ (meq/l)	Na ⁺ (meq/l)	K ⁺ (meq/l)	Mg ²⁺ (meq/l)	Ca ²⁺ (meq/l)	TDS (mg/l)	Ionic Balance	SI Calcite	SI Dolomite
#1	Turonian	Nefzaoua	26,7	7,4	422	17,0	0,6	11,6	2,0	15,9	0,5	5,3	7,4	2726	-2%	0,18	0,34
#2	Turonian	Nefzaoua	26,1	7,6	33	21,3	0,3	15,1	2,3	19,3	0,5	6,3	10,6	3729	0%	0,53	0,96
#3	Turonian	Nefzaoua	26,1	7,25	44	22,0	0,8	16,5	2,2	19,3	0,5	6,3	10,7	3801	-4%	0,16	0,21
#4	Senonian	Nefzaoua	22,9	8,05	87	11,4	0,4	4,2	1,9	9,1	0,3	3,0	4,7	1573	4%	0,68	1,25
#5	Senonian	Nefzaoua	25,0	8,11	48	35,9	0,6	7,0	1,9	26,6	0,6	5,8	7,2	3219	1%	0,82	1,68
#6	Senonian	Nefzaoua	24,1	8,01	60	16,8	0,5	4,7	1,8	12,6	0,4	3,2	4,3	1898	-1%	0,57	1,12
#7	Senonian	Nefzaoua	25,1	8,21	89	13,0	0,4	4,6	2,1	9,9	0,3	2,9	3,9	1624	-2%	0,81	1,61
#8	Senonian	Nefzaoua	25,7	8	102	13,9	0,7	5,4	2,2	10,7	0,4	3,3	4,5	1882	-1%	0,68	1,36
#9	Senonian	Nefzaoua	24,6	8,16	59	11,1	0,5	3,6	2,1	9,0	0,3	2,6	3,4	1434	1%	0,73	1,45
#10	Senonian	Nefzaoua	26,5	7,85	103	15,4	0,5	6,0	2,2	12,6	0,4	3,5	5,1	1983	0%	0,59	1,14
#11	Senonian	Nefzaoua	24,0	8,1	37	28,2	0,9	7,7	2,2	23,7	0,6	5,2	5,6	3164	-1%	0,77	1,61
#12	Senonian	Nefzaoua	24,8	8,09	88	11,5	0,5	5,0	1,8	9,7	0,3	2,7	3,6	1649	-4%	0,59	1,17
#13	Senonian	Nefzaoua	25,5	7,9	92	22,0	0,3	12,8	2,6	19,3	0,7	5,1	8,6	3294	-3%	0,81	1,5
#14	Senonian	Nefzaoua	22,0	7,9	49	3,8	1,0	1,5	1,8	3,4	0,2	1,0	1,8	648	-1%	0,22	0,29
#15	Senonian	Nefzaoua	25,1	7,75	80	20,6	0,4	17,0	1,9	20,7	0,4	6,3	11,7	3769	0%	0,6	1,06
#16	Senonian	Nefzaoua	20,4	8,28	165	14,1	0,8	13,0	2,2	15,5	0,5	6,0	7,3	2757	-1%	0,88	1,66
#17	Senonian	Nefzaoua	25,7	7,78	58	19,1	0,2	16,4	2,0	21,4	0,5	6,4	12,0	3776	3%	0,68	1,21
#18	Senonian	Nefzaoua	25,0	7,7	52	4,0	0,5	1,7	1,8	4,5	0,0	1,1	1,7	687	-3%	0,02	-0,02
#19	Senonian	Nefzaoua	24,7	7,98	271	13,3	0,5	10,5	2,0	9,2	0,3	5,6	8,8	1599	-4%	0,6	1,12
#20	Senonian	Nefzaoua	25,6	8,08	91	12,0	0,2	10,2	2,0	9,7	0,3	6,4	8,6	1723	0%	0,7	1,4
#21	Senonian	Nefzaoua	24,7	8,04	109	14,4	0,7	8,1	2,0	10,6	0,4	7,6	8,9	1719	4%	0,67	1,4
#22	Senonian	Nefzaoua	26,0	8	80	15,0	0,3	27,5	2,4	15,0	0,3	11,3	18,2	3046	-1%	-0,47	-0,96
#23	Senonian	Nefzaoua	22,3	8,09	60	77,1	0,0	24,1	2,0	52,4	1,1	12,9	36,6	7000	0%	1,06	1,77

#24	Senonian	Nefzaoua	26,5	8,06	47	12,6	0,5	14,7	2,0	11,6	0,3	7,7	9,7	2017	-2%	0,7	1,44
#25	Mio-Pliocene	Redjime Mâatoug	26,8	7,76	225	14,5	0,5	12,1	2,4	14,0	0,6	6,1	8,5	2917	3%	0,66	1,31
#26	Mio-Pliocene	Redjime Mâatoug	26,7	7,96	180	14,7	0,6	11,9	2,3	14,1	0,6	6,0	7,9	2791	2%	0,65	1,3
#27	Mio-Pliocene	Redjime Mâatoug	25,5	7,99	165	14,1	0,2	12,4	1,1	13,2	0,6	4,7	7,6	2805	-3%	0,47	0,85
#28	Mio-Pliocene	Redjime Mâatoug	26,0	7,88	164	11,3	0,5	8,5	2,2	10,9	0,5	4,5	5,2	2171	-1%	0,58	1,22
#29	Mio-Pliocene	Redjime Mâatoug	22,4	8,25	118	7,2	0,2	3,2	2,0	6,8	0,4	2,1	2,7	1167	1%	0,7	1,38
#30	Mio-Pliocene	Redjime Mâatoug	22,7	7,76	190	15,6	0,5	11,9	3,6	14,9	0,8	5,1	8,4	3070	-1%	0,78	1,43
#31	Mio-Pliocene	Redjime Mâatoug	22,8	8,48	150	7,7	0,2	3,2	2,8	7,5	0,6	1,3	3,2	1199	0%	0,86	1,7
#32	Mio-Pliocene	Tozeur	25,8	7,53	287	15,8	0,1	11,8	2,7	14,0	0,7	5,2	10,0	3028	3%	0,54	0,92
#33	Mio-Pliocene	Tozeur	29,9	7,68	279	15,2	0,2	5,8	2,1	12,8	0,4	3,0	4,3	1896	-2%	0,37	0,75
#34	Mio-Pliocene	Tozeur	28,4	7,43	85	22,3	0,2	5,3	1,9	16,4	1,4	3,5	5,2	2361	0%	0,15	0,29
#35	Mio-Pliocene	Tozeur	34,8	7,6	500	13,7	0,2	4,8	1,9	8,6	0,2	1,3	7,8	1786	3%	0,59	0,6
#36	Mio-Pliocene	Tozeur	31,7	7,33	492	18,7	0,2	10,3	2,4	9,3	0,4	4,9	13,1	2961	4%	0,51	0,76
#37	Mio-Pliocene	Tozeur	29,8	7,45	362	21,4	0,3	10,1	2,3	15,6	0,3	4,9	11,1	2853	1%	0,51	0,83
#38	Mio-Pliocene	Tozeur	31,7	7,6	98	17,1	0,2	4,9	1,8	12,9	0,4	3,0	4,1	1909	4%	0,24	0,54
#39	Mio-Pliocene	Tozeur	32,4	7,34	550	41,7	0,2	5,7	2,0	21,8	0,4	4,1	14,2	3658	3%	0,49	0,64
#40	Mio-Pliocene	Tozeur	30,4	7,16	40	34,1	0,2	15,8	2,2	26,4	0,0	7,4	11,8	4646	-2%	0,15	0,26
#41	Mio-Pliocene	Tozeur	36,2	7,29	666	21,0	0,3	10,6	2,2	16,3	0,3	4,2	11,7	3139	4%	0,43	0,61
#42	Mio-Pliocene	Tozeur	31,9	7,18	64	55,4	0,5	8,3	2,1	35,8	0,8	6,2	11,9	4851	-1%	0,23	0,36
#43	Mio-Pliocene	Tozeur	33,1	7,17	64	78,3	0,2	10,9	2,3	49,2	1,0	8,0	23,1	6290	4%	0,48	0,7
#44	Mio-Pliocene	Tozeur	28,5	7,3	312	17,5	0,3	9,6	2,4	15,4	0,1	2,5	10,3	2777	3%	0,34	0,22
#45	Mio-Pliocene	Tozeur	32,8	7,17	275	17,2	0,2	10,4	2,2	13,3	0,2	4,0	10,9	2907	4%	0,24	0,24
#46	Mio-Pliocene	Tozeur	31,1	7,14	70	17,0	0,0	14,0	2,3	14,2	0,0	5,5	10,2	3184	-2%	0,15	0,19
#47	Mio-Pliocene	Tozeur	26,8	7,42	443	19,6	0,3	9,7	2,4	16,3	0,0	4,8	7,4	2859	-1%	0,29	0,52
#48	Mio-Pliocene	Tozeur	29,3	7,33	285	15,6	0,1	4,6	2,0	16,3	0,0	2,6	3,7	1911	4%	0,64	1,21
#49	Mio-Pliocene	Tozeur	31,5	7,55	300	17,4	0,2	6,8	2,2	13,4	0,4	3,9	7,2	2321	4%	0,48	0,86
#50	Mio-Pliocene	Tozeur	30,0	8,1	320	15,3	0,3	7,5	2,3	15,5	0,5	3,4	5,6	2058	2%	0,89	1,73
#51	Mio-Pliocene	Tozeur	29,0	7,2	68	15,2	0,1	14,7	2,0	15,8	0,6	6,1	9,4	3336	1%	0,03	-0,02

#52	Mio-Pliocene	Tozeur	30,0	7,22	612	14,0	0,0	16,5	2,0	12,6	0,7	6,2	9,8	3002	-4%	0,12	0,18
#53	Mio-Pliocene	Tozeur	28,2	8,02	65	16,9	0,2	19,2	3,4	17,2	0,8	6,4	11,0	3617	-4%	1,07	2,01
#54	Mio-Pliocene	Tozeur	27,0	7,8	100	16,4	0,2	18,8	2,4	17,9	0,7	7,1	9,4	3522	-4%	-0,14	-0,29
#55	Mio-Pliocene	Tozeur	30,0	7,28	482	13,3	0,3	17,0	2,2	12,9	0,5	8,2	12,0	2138	1%	0,09	0,19
#56	Mio-Pliocene	Chott El Gharsa Plain	30,0	7,9	565	16,5	0,1	14,6	2,4	20,2	0,0	4,9	10,6	3284	3%	0,89	1,61
#57	Mio-Pliocene	Chott El Gharsa Plain	30,0	7,2	500	17,9	0,0	18,2	2,4	26,3	0,4	4,9	10,5	3961	1%	0,17	0,17
#58	Mio-Pliocene	Chott El Gharsa Plain	35,9	7,8	538	53,1	0,0	17,8	2,4	40,5	1,0	10,3	17,7	6279	4%	1	1,98
#59	Mio-Pliocene	Chott El Gharsa Plain	49,0	8	1052	21,1	0,1	15,0	2,4	25,6	0,0	4,6	10,5	3610	2%	1,13	2,12
#60	Mio-Pliocene	Chott El Gharsa Plain	39,8	8,1	1174	22,7	0,1	13,2	2,4	22,1	0,0	5,1	10,9	3392	2%	1,2	2,28
#61	Mio-Pliocene	Chott El Gharsa Plain	45,0	7,6	1052	21,1	0,1	15,5	2,3	15,8	0,4	5,7	11,4	3622	-4%	0,79	1,49
#62	Mio-Pliocene	Chott El Gharsa Plain	42,0	7,2	1164	29,2	0,1	16,1	2,5	17,9	0,5	8,1	16,9	4489	3%	0,55	0,99
#63	Mio-Pliocene	Chott El Gharsa Plain	45,8	8,19	1081	28,0	0,0	29,0	4,1	28,2	0,5	19,7	14,4	3700	1%	1,36	3,11
#64	Mio-Pliocene	Chott El Gharsa Plain	41,0	7,8	846	34,4	0,0	32,2	2,0	32,2	0,0	10,8	25,4	4500	-1%	0,88	1,61
#65	Mio-Pliocene	Chott El Gharsa Plain	30,0	7,9	660	24,0	0,0	39,4	2,8	20,0	0,5	12,6	31,0	4650	-2%	1,05	1,88
#66	Mio-Pliocene	Chott El Gharsa Plain	35,0	7,8	565	50,1	0,0	43,4	1,9	52,0	0,7	22,9	25,6	7050	3%	0,73	1,6

Ø: Hadj Ammar et al., 2014.

Table 2. Uranium isotopic data together with stable isotopes and ¹⁴C corrected age of analyzed groundwater samples of the CT aquifer.

Sample Labels	Aquifer	Basin	δ ¹⁸ O ‰ vs SMOW _θ	δ ² H ‰ vs SMOW _θ	U (ppb) γ	U error	²³⁴ U/ ²³⁸ U γ	²³⁴ U/ ²³⁸ U error	¹⁴ C activity (pmC)	¹⁴ C error	δ ¹³ C ‰ PDB	Corrected ¹⁴ C Age (Year)	Age standard deviation	A ₀ Fontes and Garnier	Uncorrected ¹⁴ C Age (Year)
#1	Turonian	Nefzaoua	-7,2	-54,5	7,927	0,014	1,386	0,002	30,2*	1,2	-7,3	1382	247,2	35,7	9898
#2	Turonian	Nefzaoua	-6,6	-50,5	7,413	0,014	1,767	0,003	na	na	na	na	na	na	na
#3	Turonian	Nefzaoua	-6,7	-50,4	6,778	0,013	1,652	0,003	11,2*	1,0	-4,5	5112	671,7	20,8	18099
#4	Senonian	Nefzaoua	-6,2	-51,7	3,334	0,007	1,732	0,006	18,1 ^γ	1,3	-5,1	2531	435,4	24,6	14130
#5	Senonian	Nefzaoua	-5,9	-44,8	4,691	0,008	1,537	0,003	na	na	na	na	na	na	na
#6	Senonian	Nefzaoua	-5,4	-45,8	3,204	0,005	1,669	0,002	10,8 ^ε	0,9	-3,6	3508	477,4	16,5	18399
#7	Senonian	Nefzaoua	-6,0	-49,9	3,600	0,006	1,740	0,002	na	na	na	na	na	na	na
#8	Senonian	Nefzaoua	-6,1	-50,9	4,304	0,007	1,808	0,003	na	na	na	na	na	na	na
#9	Senonian	Nefzaoua	-6,2	-48,7	3,071	0,005	1,695	0,002	na	na	na	na	na	na	na
#10	Senonian	Nefzaoua	-5,4	-51,5	4,186	0,008	1,879	0,002	11,6 ^ε	1,2	-5,8	7191	714,4	27,7	17808
#11	Senonian	Nefzaoua	-5,9	-46,2	5,126	0,010	1,691	0,003	na	na	na	na	na	na	na
#12	Senonian	Nefzaoua	-4,7	-46,8	5,623	0,010	1,605	0,003	9,6 ^ε	1,1	-4,6	6719	864,2	21,6	19373
#13	Senonian	Nefzaoua	-6,1	-48,8	3,577	0,006	1,776	0,003	6,7 ^γ	1,0	-5,2	10842	1243,1	24,9	22346
#14	Senonian	Nefzaoua	-5,5	-41,6	1,749	0,003	1,492	0,002	na	na	na	na	na	na	na
#15	Senonian	Nefzaoua	-6,5	-49,0	8,121	0,016	1,833	0,003	na	na	na	na	na	na	na

#16	Senonian	Nefzaoua	-6,8	-51,1	5,978	0,014	1,518	0,003	17,9 ^Y	2,0	-4,2	1055	462,3	20,3	14222
#17	Senonian	Nefzaoua	-6,9	-51,0	7,316	0,014	1,732	0,002	6,9 ^Y	1,2	-6,2	12157	1206,5	30,0	22103
#18	Senonian	Nefzaoua	-5,7	-45,2	1,981	0,003	1,134	0,002	na	na	na	na	na	na	na
#19	Senonian	Nefzaoua	-6,2	-53,9	na	na	na	na	17,7 ^Y	1,1	-4,8	2043	467,5	22,7	14315
#20	Senonian	Nefzaoua	-6,2	-50,7	na	na	na	na	8,8 ^Y	1,0	-7,9	12261	943,4	38,8	20092
#21	Senonian	Nefzaoua	-6,1	-48,6	na	na	na	na	12,4 ^Y	1,1	-4,6	5135	668,1	23,1	17257
#22	Senonian	Nefzaoua	-6,3	-50,9	na	na	na	na	na	na	na	na	na	na	na
#23	Senonian	Nefzaoua	-6,0	-49,5	na	na	na	na	20,7 ^Y	1,3	-6,5	3497	505,9	31,6	13021
#24	Senonian	Nefzaoua	-5,7	-51,3	na	na	na	na	16,4 ^Y	1,4	-6,8	5787	613,4	33,0	14946
#25	Mio-Pliocene	Redjime Mâatoug	-5,0	-47,8	5,071	0,009	3,140	0,004	na	na	na	na	na	na	na
#26	Mio-Pliocene	Redjime Mâatoug	-5,6	-49,1	4,913	0,009	2,831	0,003	na	na	na	na	na	na	na
#27	Mio-Pliocene	Redjime Mâatoug	-5,1	-49,4	3,828	0,006	2,704	0,003	na	na	na	na	na	na	na
#28	Mio-Pliocene	Redjime Mâatoug	-4,1	-44,4	4,324	0,007	2,705	0,003	10,3 ^Y	1,5	-7,7	10713	940,6	37,6	18791
#29	Mio-Pliocene	Redjime Mâatoug	-5,6	-53,5	2,667	0,004	2,134	0,002	6,9 ^E	1,0	-5,9	11766	1890,6	28,6	22103
#30	Mio-Pliocene	Redjime Mâatoug	-4,6	-46,7	4,279	0,007	2,805	0,003	na	na	na	na	na	na	na
#31	Mio-Pliocene	Redjime Mâatoug	-4,9	-45,5	2,176	0,004	1,833	0,003	7,1 ^Y	0,9	-7,4	13509	1172,1	36,4	21867
#32	Mio-Pliocene	Tozeur	-5,1	-40,1	3,942	0,008	2,685	0,005	1,75 ^Y	1	-6,8	24252	5655,5	32,9	33445
#33	Mio-Pliocene	Tozeur	-4,4	-45,1	3,059	0,006	2,237	0,006	na	na	na	na	na	na	na
#34	Mio-Pliocene	Tozeur	-5,9	-50,9	3,833	0,008	2,365	0,004	na	na	na	na	na	na	na

#35	Mio-Pliocene	Tozeur	-6,1	-34,8	3,337	0,007	2,361	0,007	na	na	na	na	na	na	na
#36	Mio-Pliocene	Tozeur	-5,6	-39,8	4,638	0,012	3,177	0,009	na	na	na	na	na	na	na
#37	Mio-Pliocene	Tozeur	-4,8	-45,8	5,641	0,013	2,584	0,006	na	na	na	na	na	na	na
#38	Mio-Pliocene	Tozeur	-5,5	-40,2	3,034	0,007	2,162	0,005	2,1 ^γ	1	-7,0	22921	5407,7	33,6	31937
#39	Mio-Pliocene	Tozeur	-5,9	-51,7	3,644	0,007	2,787	0,004	na	na	na	na	na	na	na
#40	Mio-Pliocene	Tozeur	-7,1	-53,1	7,323	0,017	1,798	0,004	3,68 ^γ	1,2	-10,8	22142	3329,9	53,6	27300
#41	Mio-Pliocene	Tozeur	-5,3	-46,2	5,529	0,014	2,218	0,007	na	na	na	na	na	na	na
#42	Mio-Pliocene	Tozeur	-5,6	-50,7	5,592	0,013	2,314	0,007	na	na	na	na	na	na	na
#43	Mio-Pliocene	Tozeur	-6,4	-48,5	5,289	0,050	2,452	0,067	11,5 ^γ	0,9	-5,7	6968	922,3	26,7	17880
#44	Mio-Pliocene	Tozeur	-4,8	-45,4	4,374	0,011	2,610	0,008	3,2 ^γ	0,8	-6,4	18636	3066,6	30,5	28455
#45	Mio-Pliocene	Tozeur	-5,2	-48,1	6,979	0,017	2,055	0,006	na	na	na	na	na	na	na
#46	Mio-Pliocene	Tozeur	-5,6	-44,3	4,652	0,011	2,852	0,007	na	na	na	na	na	na	na
#47	Mio-Pliocene	Tozeur	-4,8	-46,4	4,379	0,009	2,782	0,005	5,8 ^γ	1,2	-7,5	15192	2193,2	36,4	23539
#48	Mio-Pliocene	Tozeur	-4,8	-44,9	2,713	0,006	2,169	0,006	na	na	na	na	na	na	na
#49	Mio-Pliocene	Tozeur	-5,9	-46,8	6,639	0,019	2,246	0,009	na	na	na	na	na	na	na
#50	Mio-Pliocene	Tozeur	-4,3	-42,0	3,627	0,009	2,645	0,008	18,3 ^γ	1,0	-8,3	6645	524,4	40,9	14040
#51	Mio-Pliocene	Tozeur	-5,6	-49,6	5,714	0,013	2,833	0,006	17,8 ^γ	1,6	-6,1	4103	933,3	29,2	14269
#52	Mio-Pliocene	Tozeur	-6,2	-52,5	4,319	0,010	3,121	0,007	na	na	na	na	na	na	na
#53	Mio-Pliocene	Tozeur	-4,4	-45,5	19,530	0,051	2,491	0,007	na	na	na	na	na	na	na

#54	Mio-Pliocene	Tozeur	-5,3	-45,9	13,228	0,020	2,452	0,007	na	na	na	na	na	na	na
#55	Mio-Pliocene	Tozeur	-4,2	-42,8	na	na	na	na	na	na	na	na	na	na	na
#56	Mio-Pliocene	Chott El Gharsa Plain	-7,8	-54,9	7,281	0,014	1,641	0,004	24,9 ^t	2,0	-7,9	3338	831,1	38,8	11168
#57	Mio-Pliocene	Chott El Gharsa Plain	-6,9	-51,5	9,165	0,017	1,340	0,003	na	na	na	na	na	na	na
#58	Mio-Pliocene	Chott El Gharsa Plain	-6,4	-49,5	7,316	0,014	2,786	0,006	na	na	na	na	na	na	na
#59	Mio-Pliocene	Chott El Gharsa Plain	-7,4	-56,4	5,296	0,011	1,572	0,005	7,3 ^t	0,8	-6,4	12365	1129,6	36,6	20676
#60	Mio-Pliocene	Chott El Gharsa Plain	-7,7	-52,9	6,024	0,011	1,654	0,004	8,4 ^t	0,5	-8,2	6453	694,3	40,2	13995
#61	Mio-Pliocene	Chott El Gharsa Plain	-7,6	-57,4	5,920	0,011	1,611	0,005	6,3 ^t	0,5	-8,7	15806	1170,2	42,6	22855
#62	Mio-Pliocene	Chott El Gharsa Plain	-7,5	-54,9	10,328	0,022	1,408	0,006	7,5 ^t	1,0	-8,8	14374	1439,6	42,7	21414
#63	Mio-Pliocene	Chott El Gharsa Plain	-5,8	-55,7	na	na	na	na	17,1 ^t	1,4	-7,6	6320	776,4	36,7	14600
#64	Mio-Pliocene	Chott El Gharsa Plain	-7,3	-51,2	na	na	na	na	9,2 ^t	1,6	-8,6	8477	590,4	42,2	15601
#65	Mio-Pliocene	Chott El Gharsa Plain	-7,7	-51,0	na	na	na	na	18 ^t	1,0	-7,5	5882	710,3	36,7	14176
#66	Mio-Pliocene	Chott El Gharsa Plain	-7,1	-49,0	na	na	na	na	6,5 ^t	1,2	-6,4	12797	1675,9	30,6	22597