U Isotope Systematics of Groundwaters from the Triassic Aquifer of the Northeastern Paris Basin and of the Rhine Graben, France

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U isotope systematics of groundwaters from the Triassic aquifer of the northeastern Paris Basin and of the Rhine Graben, France.

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Abstract

Groundwaters and spring waters from the French Lower Triassic Sandstones (LTS) and the Muschelkalk aquifers have been measured for U concentrations and activity ratios. All are clearly enriched in ^{234}U, independently of the stratigraphic level and the geographic region. Attempts have been made to derive flow rates in the case of the northeastern Paris Basin, indicating that, although the flow rate is quite low (~ 0.05 m/y) , LTS groundwaters cannot be considered as completely stagnant in this area.

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Keywords: Groundwater; Uranium; Trias; Sandstones; Paris basin; Rhine graben

1. Introduction

The Lower Triassic (Buntsandstein) Sandstone (LTS) aquifer is the deepest of the Paris Basin, outcropping at its northeastern edge. It is, at present, clearly overexploited and its management becomes very critical, since it is the most important resource for drinking water in this area (Lorraine). Furthermore, LTS waters are extensively used for industrial processes, namely as process waters for the exploitation of bottled mineral waters coming from the overlying Muschelkalk aquifer : Vittel®, Contrex®, Hépar®, … Finally, they are used for irrigation, and the aquifer has been suggested to constitute an appropriate reservoir for CO_2 storage. LTS crop also out in the Rhine graben. In this area, formation fluids are either thermal waters or brines used for geothermal energy production.

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2. Sampling and analytical techniques

A number of geochemical and isotopic studies are available, both for the LTS aquifer of Lorraine[1-5] and geothermal fluids from the Rhine graben[6-8]. Here we present uranium concentrations and isotopic ($^{234}U/^{238}U$) data obtained on groundwaters from the Lorraine area and on thermal spring waters from the French side of the Rhine valley (Alsace). In addition, waters from different overlying aquifers have been measured for comparison : three water samples coming from the overlying Muschelkalk aquifer : a groundwater and a springwater from the Lorraine region and a spring water from the Alsace region, four spring waters from the Pliocene aquifer of the Rhine valley and finally the thermal water of Baden-Baden in Germany, whose origin remains unknown, either coming from the crystalline basement and/or carboniferous rocks[9] or from sub-surface formations[10]. Sample location is given in Figure 1.

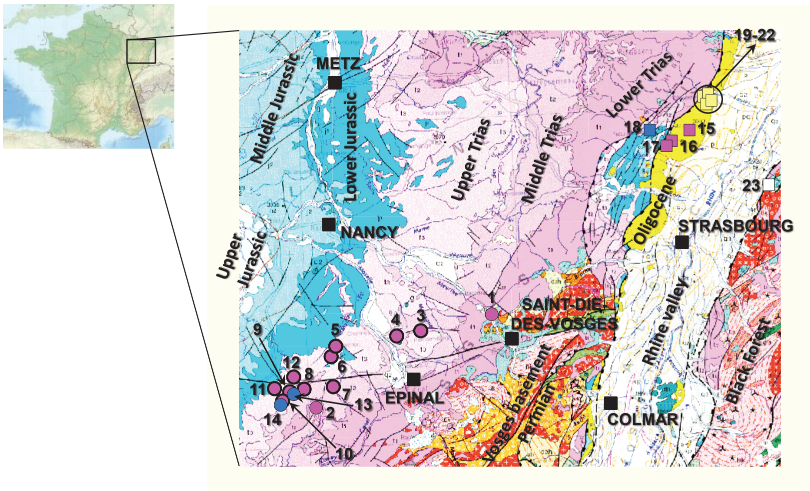

Fig. 1. Geological map of northeastern France and the Rhine valley, showing sample location. The legend is given in Table 1 (below).

For Lorraine waters, five uranium concentrations were determined by Q-ICP-MS. The other uranium contents were measured by MC-ICP-MS using the isotope dilution technique, together with uranium activity ratios after chemical separation. Waters from the Rhine valley were all measured by alpha spectrometry, both for concentrations (using the isotope dilution technique) and activity ratios, resulting in larger uncertainties. Results are reported in Table 1.

3. Results and discussion

3.1. Lorraine (northeastern Paris basin)

The two groundwaters recovered from the unconfined part of the LTS aquifer (waters 1 and 2, Tab. 1) display very high uranium activity ratios and, conversely, low U contents. ^{234}U enrichment results very probably from alpha recoil processes during water percolation within the soil. Waters 3 and 4 from the confined LTS aquifer have higher U contents, but their activity ratios are far lower. For the water from Rehaincourt (n° 4, Tab. 1), ^{14}C data[5] indicate very young ages, which could be explained by an input of recent surface waters, probably through drainage processes from overlying aquifers.

The two waters from the Mirecourt area (waters 5 and 6, Fig. 1, Tab. 1) have U characteristics that are very different, showing important heterogeneities of the groundwater flow. The first one displays a low activity ratio, close to secular equilibrium, whereas the other is largely enriched in ^{234}U. The Poussay groundwater (n° 5) has also a high U content and its ^{14}C activity has been found to be very low (< 2.5 pCM)[5]. This suggest that this water could be considered as fossil.

Sample	Aquifer	U (pg/g)	Erreur	($^{234}U/^{238}U$)	Erreur
NE Paris Basin (Lorraine)					
1 Etival-Clairefontaine*	LTS unconfined	190	10	2.984	0.008
2 Relanges	LTS unconfined	88.5	0.1	4.744	0.022
3 Saint-Genest*	LTS confined	2200	110	1.436	0.004
4 Rehaincourt*	LTS confined	1420	71	1.343	0.004
5 Poussay - Val d'Arol*	LTS confined	26460	1323	1.060	0.002
6 Mirecourt	LTS confined	6265	61	2.983	0.023
7 Valfroicourt	LTS confined	2200	1	3.520	0.016
8 Vittel	LTS confined	2800	4	3.152	0.009
9 Frênes	LTS confined	1683	2	3.519	0.020
10 Suriauville	LTS confined	758	1	5.307	0.021
11 Bulgnéville	LTS confined	437	1	7.198	0.029
12 Norroy	LTS confined	730	1	7.246	0.016
13 Contrex Great Source®	Muschelkalk	916	1	2.613	0.010
14 Le Bon Pré	Muschelkalk	1488	1	1.318	0.008
Rhine graben					
15 Merkwiller Les Hélions**	LTS	18.9	0.1	5.500	0.500
16 Morsbronn Arbogast**	LTS	615	3	6.342	0.056
17 Morsbronn Cuirassiers**	LTS	629	3	6.343	0.038
18 Niederbronn Romaine**	Muschelkalk	5524	40	3.530	0.009
19 Wissembourg "A"**	Pliocene	385	4	1.590	0.140
20 Wissembourg "B"**	Pliocene	177	1	1.764	0.058
21 Wissembourg "D"**	Pliocene	1248	12	1.760	0.014
22 Wissembourg "E"**	Pliocene	792	5	1.202	0.015
23 Baden-Baden**	Unknown	297	1	0.901	0.018

Tab. 1. U concentrations and activity ratios of the analyzed water. The single asterisk refers to water for which U concentration has been measured by Q-ICP-MS, and the double asterisk to those analyzed by alpha spectrometry for both parameters. The symbols used in the following figures are shown for each aquifer distinguishing confined and unconfined parts for LTS.

Groundwaters from the Vittel area (West of Epinal, Fig. 1) have very high U activity ratios (Tab. 1), roughly increasing along the flowpaths in the confined part of the aquifer (Fig. 2). The highest ratios are found west (waters 11 and 12), corresponding to lowest water levels. In contrast, U contents are found to decrease slightly from east to west (Fig 3). It has to be noticed that the lower activity ratios measured in water 9 (Tab. 1, Fig. 2) reflect very likely drainage processes from the overlying Muschelkalk aquifer[5].

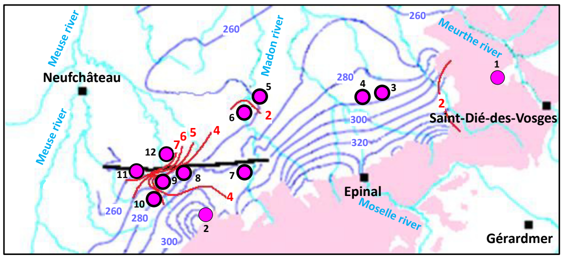

Fig. 2. Schematic map showing the piezometric levels together with U activity ratios measured in LTS groundwaters.

Such an increase of activity ratios with decreasing U contents seems to be related to the transition from the unconfined to the confined part of the LTS aquifer. This process is illustrated by the trend "1" in Fig. 3. As LTS brines located ~ 60 km northwest of waters 11 and 12, towards the center of the Paris basin, display a much lower activity ratio (1.36)[3], and also a much lower U content, indicating that there is no U input during groundwater flow, the flowing rate has been tentatively calculated using a simple downflow-return-to-equilbrium model[11] (trend "2" in Fig. 3). It is found to be in the range of 0.05 m/y, at least one order of magnitude lower than those derived for the "upstream" part of the LST aquifer[1,12]. However, this would indicate that groundwaters are not completely stagnant in the studied area.

Waters from the overlying Muschelkalk aquifer display also activity ratios higher than the secular equilibrium, but lower than those of groundwaters of the LTS aquifer (Tab. 1), water 14 (Tab. 1) giving evidence of the influence of surface waters)[5].

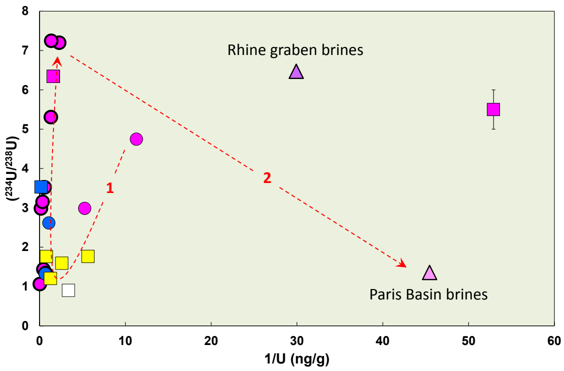

Fig. 3. U activity ratios vs. 1/U concentrations diagram for the analyzed waters. The arrow "2" is only indicative and does not represent model calculations (see above).

3.2. Rhine graben

LTS waters from the Rhine graben also display high uranium activity ratios, in the range of the most ^{234}U-enriched Lorraine LTS waters (Tab. 1, Fig. 3). However, in this case, activity ratios are very close to that of the Triassic brines from the Rhine graben[10] (which have thus activity ratios completely different from those of the Paris basin, Fig. 3). U activity ratios of Muschelkalk waters of the Rhine graben and of the Paris basin (represented by water 13, the Great Source® spring water of Contrexéville) are close to each other. Finally, it should be noticed that such high activity ratios are not characteristic of the whole Rhine graben, as Cenozoic spring waters and the Baden Baden thermal water display much lower U activity ratios, close to or at equilibrium (Tab. 1, Fig. 3).

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