



Underground renewal time and mixing of the main mineral waters of Tunisia: A multi-tracer study



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ABSTRACT

Bottled waters are increasingly popular in Tunisia. Most of them come from groundwaters, thus raising the question of the long-term availability of the mineral groundwater resources. Water collected from production springs or wells of eleven mineral waters of Tunisia among the main commercial brands were analyzed for tritium (³H), helium-3, SF₆, CFCs and carbon-14. Tracer-tracer plots indicate that all of the groundwaters of our data set are a mixture of modern rainwater with older groundwater flow waters that are beyond the time-scale of the transient tracers. ¹⁴C data suggest that the age of these old waters, which constitute the main water reservoir, is of the order of radiocarbon half-life or more. Most of the studied sites have groundwater renewal times in the range 50–150 years. For the sites where tracer concentrations are the most diluted by old waters, this value logically increase to several centuries.

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1. Introduction

The rapid increase in bottled mineral groundwater production in Tunisia needed to meet consumer demand (110 millions bottles in 1995 and 1.04 billion bottles in 2015 – Office du thermalisme at www.thermalisme.nat.tn) raises the question of the long-term sustainability of this economic sector.

Renewal time of groundwater, which is clearly related to the ratio of the storage volume to the recharge rate, is an important notion regarding vulnerability and sustainability of groundwater resources. Numerical models of groundwater flow can describe accurately the hydrodynamic functioning of a given aquifer, and therefore can give stakeholders and the mineral water industry management important indications to prevent pollution and/or overexploitation of the water resource. However, they require detailed information (e.g. boundary conditions, hydraulic conductivity, porosity, etc ...) which are usually not available. As an alternative, the analysis of atmospheric transient tracer

concentrations in groundwaters can provide important basic information on their residence time and mixing.

As a development arising from tritium, which can be used as a transient tracer as a result of ³H injection into the atmosphere from the atmospheric testing of nuclear devices in the 50's and early 60's, the ³H-³He dating method offers a direct measure for the time since groundwater had its last gas exchange with the atmosphere. The ³H-³He pair is of particular interest because tritium and its daughter helium-3 are assumed to be fully conservative tracers, and also because the ³H-³He radioactive clock only relies on the in-situ ³H/³He ratio. Hence, it is independent of the history of tritium input to the aquifer (Schlosser et al., 1988, 1989; Poreda et al., 1988; Solomon and Sudicky, 1991; 1993; Stute et al., 1997).

Like tritium, ¹⁴C was also a by-product of atmospheric nuclear tests. Hence, the resultant transient spike in atmospheric ¹⁴CO₂ can also be used as a transient tracer (Stewart, 2012; Baudron et al., 2013).

The CFCs and SF₆ methods are based on the direct comparison of the groundwater tracer concentration with the concentration of these tracers in the atmosphere, and hence in groundwater recharge. Because their atmospheric concentration has changed through time, this provides the basis for travel time determination

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(Thompson and Hayes, 1979; Cook and Solomon, 1995; Busenberg and Plummer, 1992, 2000; Goody et al., 2006; Darling et al., 2012).

Each tracer has its own atmospheric input function and each tracer method relies on its own specific set of assumptions and caveats. There are significant differences in the reliability of the derived tracer ages. Some caveats are the result of hydrogeology. For CFCs and ^{14}C additional uncertainties of the infiltration conditions influence the resulting ages. Therefore the combination of several tracer methods in parallel is highly preferable to the use of a single method (Szabo et al., 1996; Beyerle et al., 1999; Plummer et al., 2001; Corcho Alvarado et al., 2005, 2007; Massmann et al., 2008; Solomon et al., 2010; Mayer et al., 2014; Kralik et al., 2014; Delbart et al., 2014; Battle-Aguilar et al., 2017). Ideally measured tracer concentrations in a groundwater can be matched simply to a particular year of recharge. However, this requires that the groundwater moves as a result of simple piston flow, that is along parallel flowlines from recharge to discharge. In reality, groundwaters are often a mixture of water of different ages due to the complexity of the hydrogeological network and/or the internal structure of the aquifer. One basic way of resolving flow processes is to plot one tracer versus another (see for instance Fig. 3 in Darling et al., 2012). On these plots, simple piston flow and binary mixing between young recharge waters and old tracer-free waters are opposite extremes of groundwater behaviour. In most cases however, groundwater flow may be more complicated and one must test a variety of modeled reservoirs connected to each other serially or in parallel with or without dead volumes, cross flow in between or bypass flow (see Ozuyurt and Bayari, 2003 and references therein).

Here we report the application of such a multi-tracer approach to a suite of mineral waters of Northern and Central Tunisia among the main commercial brands. The region is characterized by a semi-arid climate with mild wet winters and hot dry summers (monthly average temperatures vary from 10 ± 1 °C in January to 28 ± 2 °C in August), and relatively low annual rainfall in the range 300 mm–600 mm. For this study, we selected eleven bottled water production sites (Table 1), exploiting local aquifers nested in karstified limestone and/or sandstone formations (see Appendix A). Very little is known by the operators concerning the actual extent of their mineral groundwater resources and the sustainability of their business. Comparison of mineral water withdrawal rates and recharge rates estimated from precipitation figures and catchments' surface area shows that the rate of water withdrawal exceeds the present-day recharge rate. This overexploitation of the local mineral groundwater reservoirs may have serious implications in term of water quality and economic consequences for the mineral water industry. Note that the issue of groundwater

overexploitation in Tunisia is not restricted to mineral waters: Tunisia, being a semi-arid to arid country, is facing water shortage of increasing severity as a result of population growth, rising living standards and increasing water consumption by the agricultural sector (Frija et al., 2014).

The present work aims at exploring the potential of the selected suite of transient tracers (tritium/helium-3, SF₆, CFCs and radio-carbon) to gain insight into the hydrogeological characterization of these different water reservoirs.

2. Sample collection and analytical methods

The sampled sites are shown in Fig. 1, and their main characteristics (climatic conditions, elevation, ...) are summarized in Table 1. Groundwaters were analyzed for $^{18}\text{O}/^{16}\text{O}$, D/H, ^3H , ^3He , ^4He , ^{20}Ne , CFCs (F-11, F-12, F-113), SF₆, and ^{14}C . Apart from Marwa-2009 which shows a high neon supersaturation (see below), the samples were taken at the water extraction point (at the well-head for pumped waters and at the capture point for natural springs) so that contact of the water with the atmosphere can be excluded. With the exception of a few tritium/helium and ^{14}C samples taken in October 2009, all the water samples were collected in September/October 2010.

CFCs and SF₆ samples were collected in air tight stainless steel cells of 30 and 500 ml respectively and were analyzed at the University of Rennes (Labasque et al., 2006; Ayraud et al., 2008). The CFC and SF₆ concentrations were determined by Purge and Trap (PT) extraction and analysed with a gas chromatograph equipped with an electron capture detector (GC/ECD). The uncertainty is 3% for water equilibrated with the present-day atmosphere for CFC and 5% for SF₆ (see Table 2).

Noble gas samples (He, Ne) were collected using standard refrigeration grade 3/8" copper tubes equipped with metal clamps at both ends. ^3H samples were collected in 500 ml pyrex bottle, baked at 75 °C and pre-filled with argon. The tritium and noble gas isotopic analyses (^3He , ^4He , ^{20}Ne) were carried out at LSCE (CEA-Saclay) with a MAP-215 mass spectrometer using standard procedures. Helium and neon dissolved in the water samples were first extracted under vacuum into sealed glass tubes. The accuracy is $\pm 0.8\%$ for helium and neon concentrations, and $\pm 0.4\%$ for the $^3\text{He}/^4\text{He}$ ratio. Water for tritium determination was degassed and stored during 9–12 months to allow for ^3He ingrowth. Uncertainty on tritium determination is given in Table 2. Technical details concerning laboratory treatments and analytical methods are available in Jean-Baptiste et al. 1992, 2010.

Carbon-14 samples were taken in 500 ml stainless steel cells and analyzed by accelerator mass spectrometry at the French

Table 1
Summary of the main relevant parameters for the sampled sites.

Site	Geographic Zone	Catchment type	Well depth (m)	Year of commissioning	Altitude of the site (m)	Average temperature (°C)		Average precipitation (mm)			Recharge temperature (°C) (precipitation weighed)	pH	HCO ₃ (mmol/l)
						January	July	January	July	Annual			
Marwa	Bizerte	well	72	1993	300	11,3	25,2	77	3	558	13,1	7,2	4,17
Safia (Ain Mizeb)	El Kef	surface	0	1968	880	7,1	26,5	65	9	509	12,9	7,3	3,87
Safia (Ain Ksiba)	El Kef	well	62	1992	900	7,1	26,5	65	9	509	12,7	7,0	3,71
Dima	El Kef	well	~50	2009	750	7,1	26,5	65	9	509	13,7	7,5	4,04
Cristaline	Zaghouan	well	187	2003	200	9,6	27,0	68	5	496	15,2	7,4	3,79
Aqualine	Zaghouan	well	96	2006	200	9,6	27,0	68	5	496	15,2	7,6	4,40
Ain Garci	Zaghouan	surface	0	1900	110	9,6	27,0	68	5	496	15,2	6,6	19,84
Sabrine	Kairouan	well	154	1991	106	11,7	29,1	23	5	306	17,6	7,5	3,80
Fourat	Kairouan	well	~150	2003	400	11,7	29,1	23	5	306	16,1	7,2	4,80
Hayet	Kairouan	well	226	1996	420	11,7	29,1	23	5	306	16,0	7,5	2,15
Jannet	Kairouan	well	360	2002	400	11,7	29,1	23	5	306	16,1	7,6	3,75

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