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Assessments of the natural radioactivity levels in spring sources of Djelfa South of Algeria

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Abstract

Since the genesis of the earth the Radioactivity is present in the all environment compartments (Air, Soils, waters and vegetations). The level of radioactivity is depending to the geological characteristics and the nature of rock (Volcanic or sedimentary rocks). The paper aim to determine a reference level of natural radioactivity presents in groundwater by using a portable Geiger Muller counter. 8 samples of groundwater were collected. Water samples were acidified and evaporated with hotplate at a constant temperature (60°) until obtaining a small volume (1 liter). Conditioned samples were measured with a Portable Geiger Muller counter (10h). The calculated radioactivity was vary from 0,17Bq/l to 0,19Bq/l and the equivalent dose was vary from 20µS/year to 29µS/year. These results indicate that's groundwater samples have a low radioactivity and are safe for human consumption.

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Introduction

Radioactivity is present in the environment since the creation of the earth; it is the result of the unstable state of the nuclei. The instable state of atoms allows emissions of heavy particles (Alpha, beta) and electromagnetic emissions (Gamma). In the course of research aimed at deepening the phenomenon of X-ray emission discovered in 1895 by Röntgen, Henri Becquerel in March 1896 discovered a new invisible and penetrating radiation emitted spontaneously by uranium (Choppin, 1964). The photo film in (Fig. 1) shows the effect of the Pechblende mineral on the photographic film (The experience was down in dark chamber) (Raffet and Charles, 1909).

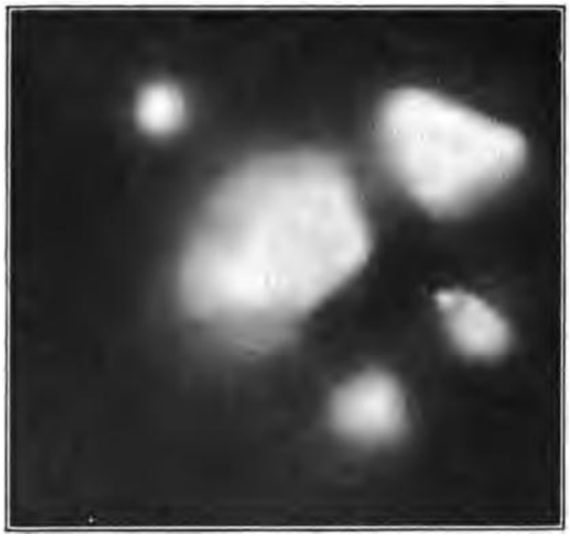

Fig. 1. Action of Pitchblende on a photographic plate Beta and Gamma rays (Raffet and Charles, 1909).

In the environment and especially in water, the radioactivity present is due to direct contact with the rocks, the water will be loaded with chemical elements to obtain its mineralization which is responsible of the presence of the natural radioisotopes (Uranium 238, Thorium 231, Potassium 40K, lead 210Pb, 212Pb, 214Pb, and 214Bi). Variation of radioactivity is depending to the nature of rocks to the depth to the surface and to the time of turnover of waters and to the geochemical process Water-Rock (Nuccetelli *et al.*, 2012). All these interactions influence the level of radioactivity. Its values arise in some thermal waters. The higher values are caused by the presence of high radioactive gaz like (Radon, 222Rn) and to the high content of 40K. Understanding the natural radioactivity allow researcher to identify waters that can threaten the human health if the cumulative absorbed dose from some radioactive water exceeds the international standards. Terrigenous radionuclide's exists in the Earth's crust since its creation. These radioelements include the parent nuclides of the three natural radioactive families 235U, 238U and 232T and nuclides devoid of radioactive progeny in addition to the Potassium 40K. The successive radioactive decays are carried out via α and γ or β emitters until a Stable isotope of lead. The main terrigenous radionuclides are shown in Table (I); particularly noteworthy is the abundance of 40 K. And 87Rb (Adloff, 1976).

Table 1. Terrigenous Radionucleides (Eisenbud, 1973).

Radionuclide	Decay (year)
^{40}K	$1{,}26 .10^{9}$
^{50}V	6.10^{15}
^{87}Rb	$1{,}8. 10^{10}$
^{115}In	$6 .10^{14}$
^{123}Te	$1{,}2 .10^{13}$
^{142}Ce	$> 5. 10^{16}$
^{144}Nd	$>2{,}4 .10^{15}$
^{148}Sm	$>2 .10^{14}$
^{146}Sm	$>1 .10^{15}$
^{152}Cd	$1{,}1.10^{14}$
^{204}Pb	$1{,}4 .10^{17}$
^{209}Bi	$\geq 2 .10^{18}$
^{232}Th	$1{,}40 .10^{10}$
^{235}U	$7{,}04. 10^{8}$
^{238}U	$4{,}45 .10^{9}$

In addition, the natural radioactivity was determinate generally by using a gamma ray spectroscopy, this nuclear technique has founded on the transformation of the emitted particles' to an electric pulses that can treated with a multichannel analyzer (MCA). This technique give the advantage that can determinate the quantitative and qualitative radioactivity of waters samples and all solid or liquid samples.

Furthermore, water quality has given a great importance in the environmental studies because of its daily uses for human consumption (Armani, 2002). All drinking water in Djelfa city (Algeria) originates from groundwater and springs sources. This study aims to determinate a reference level of radioactivity and to calculate the average daily absorbed dose for water consumer.

We focus only to determinate the quantitative levels of radioactivity without identifying the radioelement's present in water. This study helps us to calculate the specific radioactivity (Bq/liter) and to understand the radioactive quality of water in matter of citizens' health.

Material and methods

Study Area

The Djelfa basin, a tributary of Oued Melah is located between the Tellian and the Saharian atlas which has been prone to recurrent droughts in the last two decades. The basin is 1300 square kilometers, and lies between 2° E to 5° E longitudes and 33° N to 35° N latitudes. The basin's topography is frequent by Rough terrain. The altitude varies between 1568m in the upstream to 912 m in the downstream (Fig. 1). The main livelihood opportunities for rural communities in the Djelfa region are agriculture and livestock rearing. The region of Djelfa is mostly dependent on rainfall. Aquifer levels have been falling over the years because of overexploitation and decreases of groundwater recharge (Chibane *et al.*, 2015).

Samples collection and treatment

Water samples were collected from three wells and spring sources form Djelfa in two periods in the year 2015/2016. The locations are shown in Fig. 1 and Table 2.

Table 2. Samples location.

Id	Name	X [masl]	Y[masl]	Z[masl]
1	Djelfa Turonien	523 967	3 839 167	1 161
2	Rocher de sel	508 161	3 854 497	966
3	Hammam Echaref	483 993	3 828 369	950

Ten liters of water were collected in polyethylene containers. To avoid absorption of radionuclide's on the walls of the containers the water samples were acidified with nitric acid (Lydie & Nemba, 2008). In laboratory Water samples were evaporated under a constant temperature 60°C until we have obtained a small volume (about 1liter). The prepared samples was measured in real time with Geiger Muller counter during 12 hours continuously, data was saved directly via a computer interface related the counter by a standard wireless technology (Bluetooth). Recorded Data after measures will be treated separately to calculate the specific activities of water samples.

Calcul of the specific radioactivity

The activity of a sample given in Becquerel (Bq) or Curie was defined as the number of disintegration of a radioactive population per unit of time (Eichholz and Poston, 1998). Determination of the environmental radioactivity that directly and indirectly threatens human health is a difficult and expensive procedure (Yeşilkanat and Kobya, 2015).

The specific radioactivity was calculated by using the equation (Eq. 1) with taking into account the efficiency of the detector and the background radiation and the geometry of samples (Mass or volume).

$A(t) = \frac{N \pm \sqrt{N}}{\varepsilon . T . G}$ (eq.1)

Where

A (t): specific activity in Bq/l or Bq/kg

ε: Efficiency of the detector

N: Total count

Results and discussion

The geochemical characteristics of the groundwater sample were shown in the Table-3 and by the Piper and Schöeller –Berkaloff charts that allow to us to identify the geochemical classification of these waters. The Turonian is SO4-Ca type; HamamCharef :Na-K-Ca, Schöeller-Berkaloff chart allow us to identify three groups of water. The first group Ca-SO which correspond to the Turonian aquifer (this group is reach in the calcium and sulfate which they tack origin from the dissolution of Gypsum rock. The second group is Ca-Na-K type which correspond to the Hamam Echaref sample, the nature of this water indicate the abundance of the Calcium and sodium which signify a long contact with bicarbonate rock. In the graphic of Fig. 5 we report the variation of specific radioactivity measured for water samples. Results of counting are reported in table 4.

Table 3. Average physicochemical characteristics of the groundwater samples.

Id	Nom	T	pH	CE	Eh	HCO_3	Cl	NO_3	SO_4	Na	K	Mg	Ca
1	Turonian	20,6	7,77	2030	-12	414,80	197,12	2,24	378,80	92,50	4,20	16,60	297,30
2	Rocher de sel	17	8,36	914	-30	241,50	69,48	0	90,00	88,00	2,90	11,85	34,00
3	Charef	40	7,35	1643	-21	298,90	168,75	2,7	155,60	114,00	6,20	30,83	105,61

Table 4. Results of measure of specific radioactivity given in Bq/l.

Sample	Date	Time second	Count	Efficiency	Activity	Specific activity
Charef (Ch1)	08/10/2016	35012	6122,61	0,10	1,748719	0,17487
Turonian Oct-16(T1)	08/10/2016	35314	6325,36	0,10	1,791176	0,17912
Charef (Ch2)	26/03/2016	37376	7101,84	0,10	1,900106	0,19001
Charef (Ch3)	20/10/2015	37376	6655,12	0,10	1,780586	0,17806
Rocher de sel (RS1)	08/10/2016	37376	6694,75	0,10	1,79119	0,17912
Rocher de sel (RS2)	26/03/2016	37365	6432,44	0,10	1,721516	0,17215
Rocher de sel (RS3)	20/10/2015	48288	8553,12	0,10	1,771272	0,17713
Rocher de sel (RS4)	14/02/2016	36421	6497,40	0,10	1,78397	0,17840
Max			8553,1164		1,9001	0,1900
Min			6122,6147		1,7215	0,1722
Avg			6797,8299		1,7861	0,1786
Sdv			716,5328		0,0485	0,0048
Cv			0,11		0,03	0,03

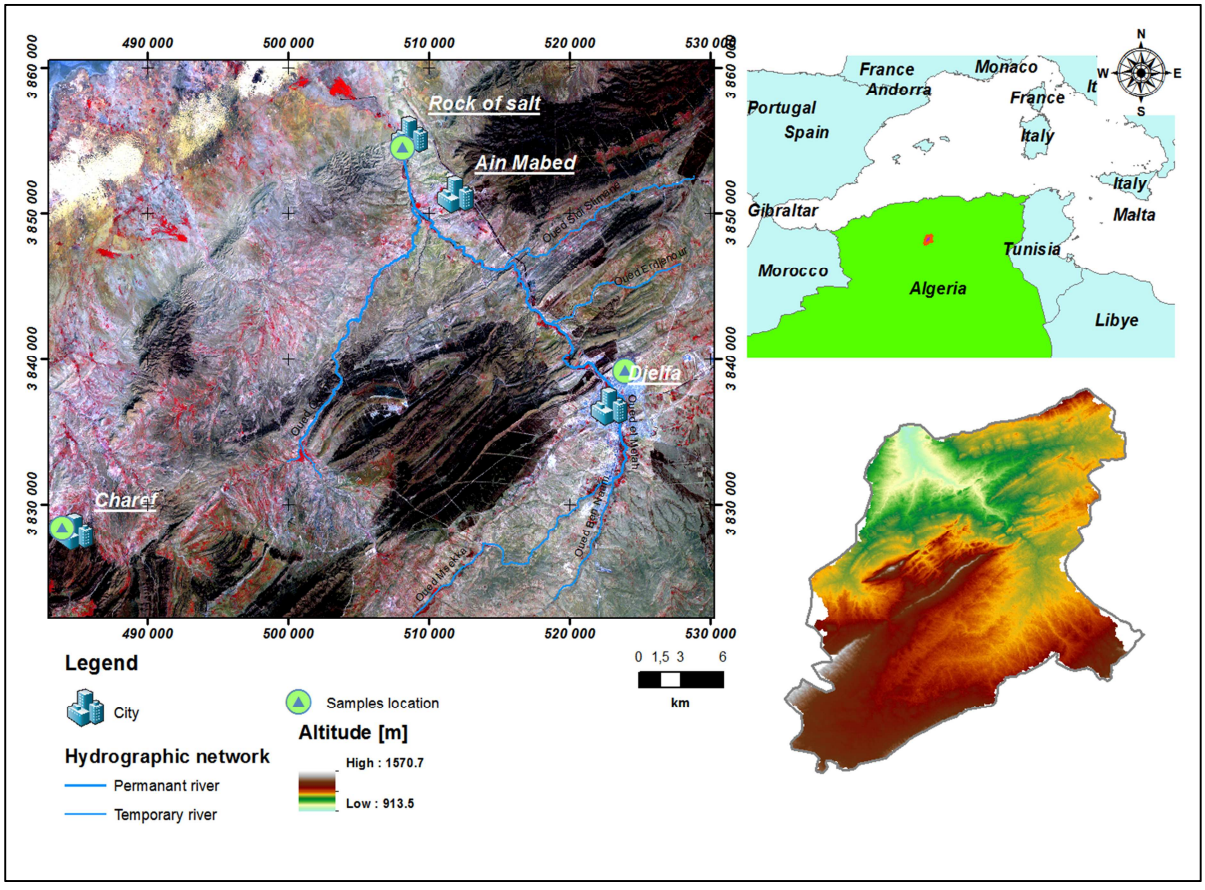

Fig. 2. Djelfa watershed and samples location.

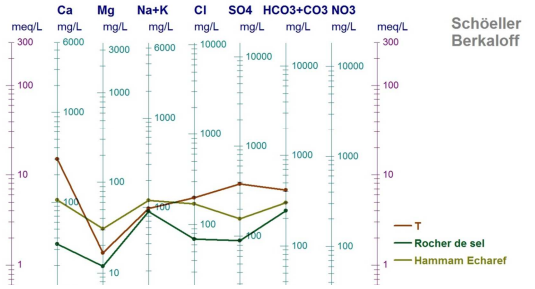

Fig. 3. Water samples classification using Schöeller-Berkaloff.

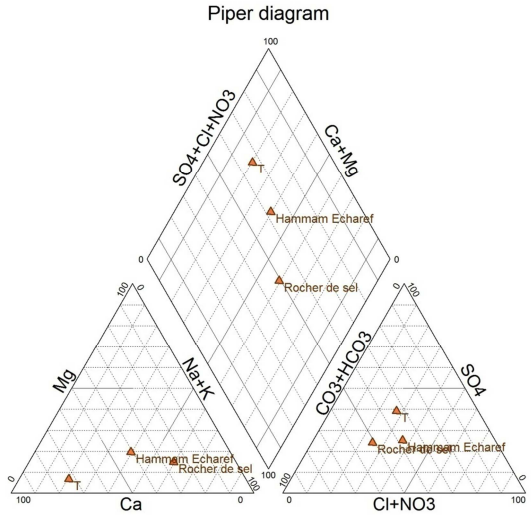

Fig. 4. Water samples classification using Piper chart.

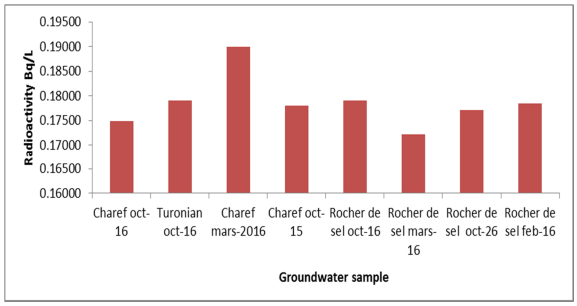

Fig. 5. Variation of average measured radioactivity for Water samples.

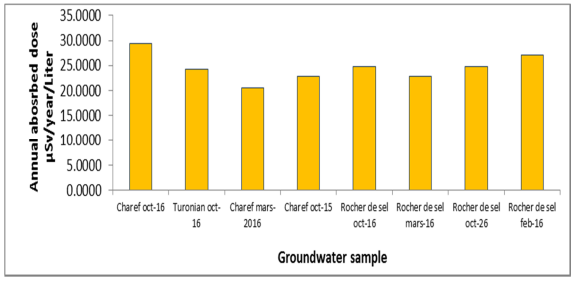

Fig. 6. Variation of average annual absorbed dose for Water samples.

The measured count was range between 6123 and 8553 count for a variable measuring duration (9 hours to 13 hours). With an efficiency of 10% the calculated radioactivity was range from 0.17Bq/l to 0.19Bq/l. The lower measured activity is corresponding to the sample of spring sources of (Rocher de sel) sampled in March, 2016. The higher is corresponding to the sample of Charef sampled also in March 2016. Analysis of the seasonal fluctuation may provide important information's about the groundwater flow. Changes observed in the groundwater samples may caused by the geochemical process and flow recharge and discharge. It reflects all the geochemical process occurred in the depth of the aquifer. The radioactivity measured it considered as low, and is safe for human consumption without any reserve (Armani, 2002). By introducing the geochemical characteristics of observed groundwater samples it appear that the measured activity it is not depend to the geochemical nature of these waters, but there are other factors that control range of radioactivity.

The measured radioactivity in some soil and rock sample is greater than in water because soil is mainly composed by high content of radionuclides with abundant percentage.

The absorption dose was determinate by the specific portable Geiger counter. Measure time is tack 10 hours to obtain the best representative results. The max value was selected for each sample to evaluate the absorbed dose in µSv/h. The total annual dose was evaluated by converting the absorbed dose given in µS/h to µS/year by introducing the daily human water consumption of these waters (Eq.2):

$$Dy = 365.d_m.WE \quad \text{(Eq.2)}$$

Where D: absorbed dose given by µSv/y/(Volume or mass unit) ; dm measured dose by Geiger counter given in µSv/h, WE: daily human water consumption.

The daily water consumption of theses water was evaluated to 3.5liter/day. The graphic in Fig. 6 and results of table-5 show the variation of absorbed data for groundwater samples.

Table 5. Results of measure of absorbed dose for the different samples.

ID	Sample	Date of sampling	Average dose µSv/h	Daily dotation Liter/day	Daily dose µSv/h	Annual dose µSv/year
1	Charef (Ch1)	08/10/2016	0,0630	3,5000	0,0805	29,3761
2	Turonian Oct-16(T1)	08/10/2016	0,0520	3,5000	0,0664	24,2470
3	Charef (Ch2)	26/03/2016	0,0440	3,5000	0,0562	20,5167
4	Charef (Ch3)	20/10/2015	0,0490	3,5000	0,0626	22,8481
5	Rocher de sel (RS1)	08/10/2016	0,0530	3,5000	0,0677	24,7132
6	Rocher de sel (RS2)	26/03/2016	0,0490	3,5000	0,0626	22,8481
7	Rocher de sel (RS3)	20/10/2015	0,0530	3,5000	0,0677	24,7132
8	Rocher de sel (RS4)	14/02/2016	0,0580	3,5000	0,0741	27,0447
Max			0,0630	3,5000	0,0805	29,376
Min			0,0440	3,5000	0,0562	20,516
Avg			0,0526	3,5000	0,0672	24,538
Sdv			0,0054	0	0,0070	2,54
Cv			0,10	0,00	0,10	0,10

The committed dose due to ingestion of inhalation of radionuclide contain in sampled groundwater's range from 20,52µSv/year to 29,37µSv/year. The origin of radionuclides in groundwater is the nature and the composition of geological deposits that contain different compositions of radionuclide's (Uranium and thorium series) in addition to the 40K that founded in abundant concentration in earth crust. The dissolution of radionuclides in water is controlled by a complicated geochemical process; the heavy radionuclides are dissolved with very slow rate. The reaction of radionuclide's in contact of water is not the same when these waters enter in reaction with major elements (Chloride, Calcium, Magnesium, Bicarbonate and the other compositions of rocks.

The concentration dioxide of Carbone, the pH and the temperature are the main factor that controls the dissolution of the radionuclides in water. In addition to the depth and the time of residency. Every time the depth is high and the time of turnover is high, the content of radioelements in water increases (Emran Eisa and al, 2015). This will give to these water their radioactivity mark and trace.

When we try to compare the results obtained in this study to the average committed dose due to ingestion and inhalation of terrestrial radionuclide's in the world (0,23mSv/y) (UNSCEAR, 1993; Amrani, 2003), we see that the contribution of these waters to the total dose considered in this work is 10,5% of worldwide average dose. The calculated effective dose for ingestion of the radionuclide's contained in mineral waters (24,53µSv/y) is insignificant compared with the value of 0,23mSv/y.

In the second way, we can conclude from these results that the groundwater of the region have low radioactivity and can't considered as dangerous for human consumption.

The radioactivity in groundwater is not depend only to the geological characteristic of the aquifer, but is depend basically to the geochemical process (Nguyen and Barbara 2009) , the time of turnover and the nature of radionuclide that enter in action with water. The pH and the CO_2 content in water allow the dissolution of the heavy isotopes like the Uranium and thorium and radium in waters. Because the neutral pH of controlled water it seem that its content of Radionuclide's is low because of these factors the measures radioactivity is low (Ljudmila and al, 2015).

Conclusion

This paper is carried out to evaluate the reference level of natural radioactivity in groundwater samples by using a new Geiger Muller counter. The specific activity was calculated in basis of the counting procedure. Results obtained show that the specific activity was range from 0,17Bq/l to 0,19Bq/l. The absorbed dose was vary between 0,021mS/year and 0,029mS/year; with an average of 0,024mS/y. radioactively these waters are safe and not dangerous for human consumption. The variation in measured radioactivity across the monitoring time can reveal some information about groundwater flow.

However the augmentations in the radioactivity level show that the residency time is important via the first waters. This is needed to be confirmed in more detailed research. Historically, waters in region are not known as radioactive water, against other region in Algeria.

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