

Uranium and radium activities measurements and calculation of effective doses in some drinking water samples in Morocco

Oum Keltoum Hakam¹, Abdelmajid Choukri¹, Aicha Abbad², Ahmed Elharfi³

¹Department of Physics, University of Ibn Tofail, Faculty of Sciences, Kenitra, Morocco

²Department of Geography, University of Ibn Tofail, Faculty of Human Sciences and Letters, Kenitra, Morocco

³Department of Chemistry, University of Ibn Tofail, Faculty of Sciences, Kenitra, Morocco

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Abstract

Purpose: As a way of prevention, we have measured the activities of uranium and radium isotopes (²³⁴U, ²³⁸U, ²²⁶Ra, ²²⁸Ra) for 30 drinking water samples collected from 11 wells, 9 springs (6 hot and 3 cold), 3 commercialised mineral water, and 7 tap water samples. **Methods:** Activities of the Ra isotopes were measured by ultra-gamma spectrometry using a low background and high efficiency well type germanium detector. The U isotopes were counted in an alpha spectrometer. **Results:** The measured Uranium and radium activities are similar to those published for other non-polluting regions of the world. Except in one commercialised gaseous water sample, and in two hot spring water samples, the calculated effective doses during one year are inferior to the reference level of 0.1 mSv/year recommended by the International Commission on Radiological Protection. **Conclusion:** These activities don't present any risk for public health in Morocco. The sparkling water of Oulmes is occasionally consumed as table water and waters of warm springs are not used as main sources of drinking water.

Keywords: Uranium; Radium; Public Health; Radioprotection; Effective Doses

Introduction

The natural radioactivity comes mainly from the primordial radio-nuclides in soils (⁴⁰K and the 4n and 4n+2 series). Further, some cosmogenic radionuclides and/or radionuclides originated from pollution can be found in natural waters used for drinking. The natural polluting radio-elements can rise from industrial wastes, geological erosion of U-bearing rocks or excessive utilization of agricultural fertilizers. Concentrations of natural radio-elements in water depend on the physico-chemical conditions and on the geological, geographical and socio-economical environment.

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has estimated that exposure to natural sources contributes more than 98% of the radiation dose to the population (excluding medical exposure). There is only a very small contribution from nuclear power production and nuclear weapons testing. The global average human exposure from natural sources is 2.4 mSv/year. There are large local variations in this exposure depending on a number of factors, such as height above sea level, the amount and type of radionuclides in the soil, and the amount taken into the body in air, food, and water. The contribution of drinking-water to the total exposure is very small and is due largely to naturally occurring radionuclides in the uranium and thorium decay

series. Levels of natural radionuclides in drinking-water may be increased by a number of human activities.

Because different types of radiation have different biological effectiveness and different organs and tissues in the body have different sensitivities to radiation, the ICRP has introduced radiation and tissue-weighting factors to provide a measure of equal effect. The sum of the doubly weighted dose received by all the tissues and organs of the body gives a measure of the total harm and is referred to as the effective dose. Moreover, radionuclides taken into the body may persist, and, in some cases, the resulting exposure may extend over many months or years. The effective dose is a measure of the total dose incurred over a lifetime following the intake of a radionuclide.

The recommended reference level of the effective dose is 0.1 mSv from 1 year's consumption of drinking-water. This reference level of dose represents less than 5% of the average effective dose attributable annually to natural background radiation. Below this reference level of dose, the drinking-water is acceptable for human consumption, and any action to reduce the radioactivity is not necessary. This recommendation does not differentiate between natural and man-made radionuclides.

Corresponding author: Oum Keltoum Hakam; Department of Physics, University of Ibn Tofail, Faculty of Sciences, Kenitra, Morocco.

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The natural radioactivity of groundwater varies greatly.¹⁻⁶ High concentrations of natural radioactive elements have been found not only in water from U mines, but also in mineral water and in some drilled wells. The knowledge of U and Ra concentrations in drinking water is important because an appreciable fraction of the absorbed U and Ra is deposited in human bone, with the corresponding contribution to the internal dose.

In this work, we describe the techniques applied for measuring U (alpha spectrometry) and Ra (gamma spectrometry) isotopes in natural water samples.⁷⁻⁸ Some results obtained from drinking water samples collected from different sources (springs, wells and tap water) are given and equivalent effective doses are calculated and compared to the recommended reference level for adult persons.

Methods and Materials

Sampling sites

The analyzed water samples belong to 3 different water sources: wells, springs (hot springs, cold springs, commercialised mineral waters), and Tap waters. **Figure 1** shows the sites of sampling. This study was undertaken in order to establish a radioactivity distribution map in various natural waters in Morocco. The regions of the cities of Fes and Errachidia are rich in warm thermal springs. Regions situated inward by the country are often affected by the drought, what explains the considerable number of wells in these regions (Khouribga, Beni Mellal and Fkih Ben Salah)

Sample preparation

Twenty liters of water are collected in a polyethylene tank and immediately slightly acidified to pH 2-3 with concentrated HNO₃. After filtration, an iron carrier (FeCl₃) (50-80 mg) and a known amount of ²³²U-²²⁸Th equilibrated spike solution are added. U and Th are coprecipitated with Fe(OH)₃ by addition of NH₄OH to pH 8 to 9 as described by Choukri and Hakam.⁷⁻⁸ A known quantity of barium in BaCl₂ form is then added. Ra isotopes are co-precipitated with BaSO₄ formed by addition of H₂SO₄ drops in the medium without significant changing of pH. Precipitation of hydroxides is achieved before the Ba and H₂SO₄ addition step to avoid co-precipitation of Th with H₂SO₄. The two precipitates are recovered by centrifugation and/or filtration.

Hydroxides containing U and Th are soluble in acid medium whereas BaSO₄ coprecipitated with Ra is not soluble in this medium. The two fractions are separated by filtration or centrifugation.

Measure of uranium

Complete mixing and oxidation are achieved by evaporating the solution to dryness. The dry residue is dissolved in 8N HCl and the hydroxides are precipitated at pH 8 to 9 with

ammonium hydroxide. In this step, Th and U are precipitated together with iron hydroxides, aluminium and other impurities. After dissolution in 8N HCl, the sample is filtered and is ready for the ion exchange procedure.⁷⁻⁸

A single resin exchange column is used twice: (1) In the first step, U and Fe are fixed on the resin in a 8N HCl medium while Th and other impurities are collected with the effluent. Fe is then eluted with 15 ml of 8N HNO₃. U is eluted with 30 ml of 0.1N HCl and evaporated to dryness, and (2) In the second step, Th is loaded on the resin bed in 30 to 40 ml of 8N HNO₃ to ensure solely the fixation of Th. The latter is eluted with 8N HCl and evaporated to dryness. The ion exchange procedure is identical to that used by Choukri *et al.*⁷ and Ku *et al.*⁹ for U and Th separation in carbonate samples with minor modifications. [**Figure 1**]

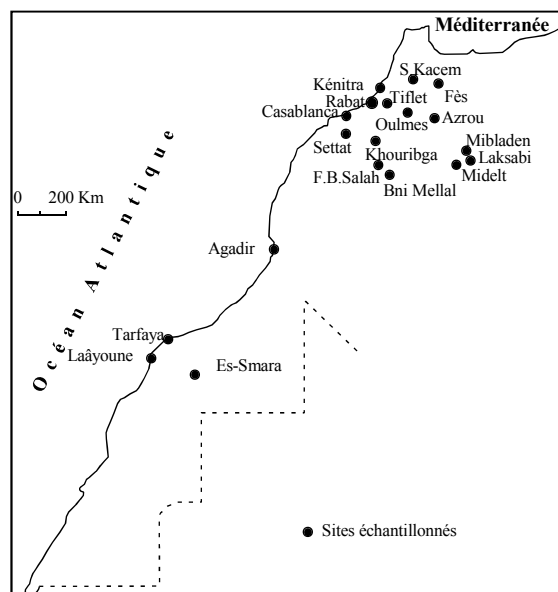


FIG. 1: Map of sampling sites

After these anionic exchange steps, Th is ready for deposition while U must be separated from residual iron. Its purification is performed by di-isopropyl ether extraction, followed by evaporation of the aqueous fraction containing U. In the last step, U and Th are extracted separately with TTA (1-(2-thenoyl)-3, 3, 3-trifluoroacetone) in toluene in a nitric media at pH 3 and 1, respectively. The two organic phases containing U and Th are evaporated onto aluminium foils and flamed to remove any trace of carbon. The thin sources are, at the end, ready for alpha spectrometry.

Determination of radium

Radium is extracted from the water samples by precipitation with BaSO₄ after removal of U and Th as described above. Schmidt and Reyss reported that this method is essentially quantitative for Ra removal.¹⁰ The (Ba-Ra) SO₄ is dried at room temperature. The obtained powder is sealed into a plastic ferrule whose form is compatible with the dimensions

of the used well Germanium detector. The gamma spectrum is measured after 20 days which is necessary to ensure radioactive equilibrium between Ra isotopes and their daughters. The efficiency of Ra extraction by BaSO₄ precipitation is obtained by calculating the weight of recovered BaSO₄ to introduced BaCl₂ (corrected by a mass factor of 1.16). Extraction efficiencies are, in most cases, in the range 75-100%. Ra activity is measured by gamma spectrometry using a 220 cm³ low-background well type gamma ray detector in the Laboratoire Souterrain de Modane (LSM-Centre National de la Recherche Scientifique (CNRS-France)/Commissariat à l'Energie Atomique).

Results and Discussion

The activities of uranium isotopes (²³⁴U, ²³⁸U) and radium isotopes (²²⁶Ra, ²²⁸Ra) and the calculated committed effective doses during one year for all analyzed samples, are given in **Table 1**. The errors quoted in this table are one sigma uncertainties due to counting statistics.

In hot springs, the activities vary between 4.92 and 173 with a mean value about 56 mBqL⁻¹ for ²³⁴U, 5.07 and 107 with a mean value about 29.6 mBqL⁻¹ for ²³⁸U, 9.10 and 122 with a mean value of 47 mBqL⁻¹ for ²²⁶Ra and between 2.84 and 16.2 with a mean value of about 7 mBqL⁻¹ for ²²⁸Ra. In well water samples ²³⁸U activities vary between 4.37 and 273 with a mean value of 85.3 mBqL⁻¹ for ²³⁴U, 4.46 and 164.8 with a mean value of 58.4 mBqL⁻¹ for ²³⁸U, 1 and 114 with a mean value of about 27 for ²²⁶Ra and between 0.6 and 51.5 with a mean value of 11.3 for ²²⁸Ra.

In Tap water samples, the activities vary between 2.85 and 58 with a mean value about 25 mBqL⁻¹ for ²³⁴U, 2.5 and 34 with a mean value about 16.24 mBqL⁻¹ for ²³⁸U, 0.46 and 45.8 with a mean value of 28.94 mBqL⁻¹ for ²²⁶Ra and between 0.4 and 30.5 with a mean value of about 10.22 mBqL⁻¹ for ²²⁸Ra. Results obtained for tap water samples did not present any anomalous value of U and Ra activity. Radioactivity in tap water depends on its source that could be rivers, dams, wells or springs.

TABLE 1: ²³⁴U, ²³⁸U, ²²⁶Ra and ²²⁸Ra activities and calculated total effective doses during one year in analyzed drinking water samples.

n°	Site	location	²³⁴ U (mBq L ⁻¹)	²³⁸ U (mBq L ⁻¹)	²²⁶ Ra (mBq L ⁻¹)	²²⁸ Ra (mBq L ⁻¹)	Total Committed Effective dose (mSV/year)
Hot springs							
1	Midelt	Moulouya	173 ± 11	107 ± 7	12.7 ± 0.1	2.84 ± 0.12	0.031
2	Mibladen	Moulouya	55 ± 5	37 ± 3	19.3 ± 0.4	6.6 ± 0.6	0.037
3	SidiHarazem	Fez	36.02 ± 2.56	5.19 ± 0.52	89.10 ± 1.20	16.20 ± 1.10	0.151
4	Ain Allah	Fez	25.70 ± 2.38	5.40 ± 0.58	30.00 ± 0.60	3.10 ± 0.10	0.050
5	Sidi Ali	Oulmes	4.92 ± 0.74	5.07 ± 0.76	9.10 ± 0.60	3.30 ± 0.60	0.016
6	OuledReguia	FB Salah	41.12 ± 2.29	17.88 ± 1.17	122.00 ± 2.10	9.60 ± 1.60	0.200
Commercialised mineral water							
7	SidiHarazem	bottle	36 ± 2.6	5.19 ± 0.5	51.9 ± 1.3	9.1 ± 1.1	0.088
8	Sidi Ali	bottle	4.9 ± 0.7	5.07 ± 0.8	9.1 ± 0.6	3.3 ± 0.6	0.016
9	Oulmès	bottle	2.4 ± 0.3	0.59 ± 0.1	1248 ± 3	66.5 ± 1.9	2.011
Cold springs							
10	Foum Anser	BeniMellal	10.62 ± 1.20	2.19 ± 0.36	10.50 ± 0.47	3.30 ± 0.60	0.019
11	AinAsserdoun	BeniMellal	15.13 ± 1.23	2.85 ± 0.32	2.45 ± 0.25	0.75 ± 0.19	0.005
12	Tirhboula	BeniMellal	22.41 ± 1.37	2.70 ± 0.24	9.05 ± 0.49	2.40 ± 0.50	0.016
Wells							
13	Mibladen	Moulouya	273 ± 25	97 ± 9	22.7 ± 0.3	8.7 ± 0.4	0.053
14	Midelt	Moulouya	98 ± 9	57 ± 5	12.9 ± 0.2	3 ± 0.2	0.027
15	F Echgag	Es-Smara	181 ± 16	114 ± 10	80 ± 0.5	26.6 ± 0.6	0.037
16	7 km-Tarfaya	Lâayoune	42 ± 4	44 ± 4	114 ± 0.8	51.5 ± 1.1	0.009
17	Settat	Settat	166.45 ± 27.10	164.80 ± 26.80	10.83 ± 0.50	17.26 ± 1.26	0.044
18	Beni Yakhlef	Khouribga	4.37 ± 0.28	4.46 ± 0.32	5.10 ± 0.30	1.00 ± 0.20	0.026
19	Ourghida	Khouribga	24.33 ± 1.98	22.53 ± 1.84	25.00 ± 1.00	5 ± 1	0.003
20	Lahmina	Khouribga	30.39 ± 2.27	26.20 ± 1.98	15.10 ± 1.40	0.60 ± 0.40	0.017
21	Beni Mellal	BeniMellal	13.19 ± 0.99	6.25 ± 0.53	1.00 ± 0.20	1.20 ± 0.30	0.014
22	Ouled Reguia	FB Salah	61.21 ± 4.17	60.60 ± 4.13	6.31 ± 0.32	4.80 ± 0.50	0.037
23	Beni Amir	FB Salah	44.12 ± 3.64	45.48 ± 3.75	5.42 ± 0.38	4.90 ± 0.60	0.009
Tap water							
24	Lâayoune	Lâayoune	41 ± 4	34 ± 3	34.0 ± 0.5	14.4 ± 0.6	0.064
25	Gwaiase	Es-Smara	58 ± 5	33 ± 3	45 ± 0.7	30.5 ± 0.9	0.090
26	Kenitra	Kenitra	2.85 ± 0.32	2.50 ± 0.30	0.46 ± 0.06	0.77 ± 0.04	0.001
27	khouribga	khouribga	21.67 ± 1.90	15.70 ± 1.41	34.60 ± 1.50	7.70 ± 1.40	0.060
28	K Tadla	K Tadla	15.06 ± 1.06	7.24 ± 0.56	40.60 ± 1.70	5.40 ± 2.20	0.068
29	FB Salah	FB Salah	24.68 ± 1.76	12.92 ± 0.98	45.80 ± 0.90	12.40 ± 1.00	0.080
30	Beni Mellal	BeniMellal	12.80 ± 1.67	8.31 ± 1.24	2.12 ± 0.29	<0.40	0.004

In cold springs, the activities are low than in hot springs. In springs, both Ra and U distributions are affected by temperature, salinity, and redox condition. High temperature tends to decrease U but to increase Ra concentrations. The aqueous chemistry of Ra may also be greatly influenced by redox state of the environment, but the effect is opposite to that for U. In geothermal sources where the temperature and the concentration of the chlorine are very elevated and the reducing potential is very weak, the $^{226}\text{Ra} / ^{238}\text{U}$ activity ratios are often very large. Zukin et al. and Herczeg et al. have found in samples of geothermal origin, the ratios in the order of 10^3 to 10^4 .¹¹⁻¹² Indeed, in a reducing medium where U is much less mobile than Ra, the disappearance of sulphates allows more important concentrations of Ra in solution.¹³

In the 3 analysed commercialised mineral water samples, the measured activities are low except in the gaseous mineral water of Oulmes where a high value of 1248 mBqL^{-1} was reached.

The risks due to the contamination of the water are essentially risks of internal irradiation as a result of ingestion of natural radioelements. To demonstrate that the annual limit of 0.1 mSv recommended for the drinking water is respected, we calculated the engaged doses which would receive the consumers from different water sources in Morocco. The dose supplied for one person via the water consumption depends not only on the quantity of radioelements ingested during one year, but also the coefficients of effective dose engaged by unity of incorporation (DPUI) recommended by the ICRP.¹⁴ (Table 2). These coefficients allow converting the activities expressed in becquerels (Bq) to dose expressed in sieverts (Sv). These values of DPUI were also taken back by the European directive.¹⁵⁻¹⁷

The calculation of the effective dose was made in our work supposing that the total quantity of ingested radioactive substances results from the consumption about 2 liters of water by a day, that is approximately 730 liters by year. The formula used for the calculation of the engaged effective dose is the following one:

$$E_T = \sum_j e_j I_j$$

where, e_j is the dose per unit intake by ingestion for adult members of the public (DPUI in Sv/Bq) by ingestion of the radioelement j ; I_j is the incorporation by ingestion of the radioelement j during the year (that is the volumic activities in becquerels by liter multiplied by 730 L).

These calculations do not consider ^{40}K , because its content in the human body is regulated and unchanging whatever is the contribution by ingestion.¹⁸ The activities of isotopes ^{210}Pb and ^{210}Po were not calculated, but estimated at a maximal equivalent activity to that of the ^{226}Ra . Considering its strong affinity for the solid phases, the thorium was not retained in the calculation of dose.¹⁹⁻²⁰

The basic formula is finally the following shape:

$$E_T = 730 (4.5 \cdot 10^{-8} \times ^{238}\text{U} + 4.9 \cdot 10^{-8} \times ^{234}\text{U} + 2.8 \cdot 10^{-7} \times ^{226}\text{Ra} + 6.9 \cdot 10^{-7} \times ^{228}\text{Ra} + 1.2 \cdot 10^{-6} \times ^{210}\text{Po} + 6.9 \cdot 10^{-7} \times ^{210}\text{Pb})$$

In radioprotection, the effect of the ingestion of radioelements is not the same as it is about a child or about an adult.²¹ The annual water consumption as well as the coefficients of effective dose engaged by unity of incorporation (DPUI) recommended by the ICRP differ according to the age of the public considered (Table 2).

TABLE 2: Dose per unit intake by ingestion for adult members of the public (DPUI)¹⁴

Category	Radionuclide	Adult	Infant
		DPUI (Sv/Bq)	DPUI (Sv/Bq)
<i>Natural uranium series</i>	Uranium-238	4.5×10^{-8}	8.0×10^{-8}
	Uranium-234	4.9×10^{-8}	8.8×10^{-8}
	Radium-226	2.8×10^{-7}	6.2×10^{-7}
	Lead-210	6.9×10^{-7}	2.2×10^{-6}
	Polonium-210	1.2×10^{-6}	4.4×10^{-6}
<i>Natural thorium series</i>	Thorium-232	2.3×10^{-7}	3.5×10^{-7}
	Radium-228	6.9×10^{-7}	3.4×10^{-6}
	Thorium-228	7.2×10^{-8}	2.2×10^{-7}

The calculated total effective doses during one year are lower than the recommended value of 0.1 mSv by the WHO for drinking water except in two hot springs and in gaseous commercialised mineral water.²² In spite of that, these waters do not present any sanitary danger for the population. The sparkling water of Oulmes is occasionally consumed as table water and waters of warm springs are not used as main sources of drinking water. Higher values were measured in China, in France, in Tunisia, in Germany and in Italy.²³⁻²⁵

Conclusion

Radiochemical analyses of 30 drinking water samples taken from 11 wells, 6 hot springs, 3 cold springs, 3 commercialised mineral water and 7 tap water show, for each source, comparable results (^{234}U , ^{238}U , ^{226}Ra and ^{228}Ra activities) to those reported in previous works for different regions in the world. The measured activities covered wide ranges. The calculated total effective doses during one year are lower than the recommended value of 0.1 mSv by the WHO for drinking water except in two hot springs and in gaseous commercialised mineral water. In spite of that, these waters do not present any sanitary danger for the population. The sparkling water of Oulmes is occasionally consumed table water and waters of warm springs are not used as main sources of drinking water.

We cannot generalize the obtained results for all drinking waters in Morocco because of the limited analyzed samples number. Although, they could give a general idea of U and Ra

radio-isotopes activity repartition in wells. springs and tap water samples which could be used to orient the next samplings in order to establish a global distribution map of radio-elements in drinking waters of Morocco.

Conflict of interest

The authors declare that they have no conflicts of interest. The authors alone are responsible for the content and writing of the paper.

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