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RESEARCH

Assessment of natural radioactivity in Moroccan bottled drinking waters using gamma spectrometry

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Abstract Radioactive elements and their impact on the environment and the food chain, including humans, are a matter of major concern, for which appropriate investigations should be performed. The priority is to examine the concentration of radioactive substances in mineral and bottled spring water. This task aims to analyze the quality of 12 conditioned mineral waters by determining their main radionuclides concentrations, such as ^{238}U, ^{232}Th, and ^{40}K. The identification and the quantification of these radionuclides are carried out by their progeny (except the ^{40}K) by using a NaI(Tl) detector coupled with a multichannel analyzer (MCA) and connected to a computer. The activity measured in all samples varied from 0.95 to 3.38 mBq.L^{-1} with an average of 1.94 mBq.L^{-1}; from 1.55 to 3.56 mBq.L^{-1} with an average of 2.46 mBq.L^{-1}; and from 200.68 to 269.19 mBq.L^{-1} with an average of 236.6 mBq.L^{-1}, for ^{238}U, ^{232}Th, and ^{40}K, respectively. To compare the combined radiological effects of radionuclides present in water, a particular factor Ra(eq) is used. This study showed that the maximum value of Ra(eq) is 27.54 mBq.L^{-1}, which is far below the activity limit of 370 mBq.year^{-1} set by the Organization of Economics and Development (OECD). Concerning the effective annual dose, the following maximums were measured: 1.61 μSv.year^{-1}, 1.133 μSv.year^{-1}, and 0.925 μSv.year^{-1} for infants, children, and adults, respectively. These values are even smaller than the dose recommended by the WHO which is 100 μSv.year^{-1}. Regarding the excess lifetime cancer risk index, a maximum of 5.63×10^{-6} is found. This index value is still less than that proposed by James, namely 2.5×10^{-3}. Thus, the quality of the studied samples respects the radiological international safety and health limits.

Keywords Gamma spectrometry · NaI(Tl) · Bottled drinking water · Natural radioactivity · Dose assessment

Introduction

The consumption of water is essential because it allows, above all, to maintain the volume of water essential for the proper functioning of our body. The radiological risk caused by the daily ingestion of radionuclides present in drinking water can increase radiological risk. This water contains natural radionuclides such as the ^{238}U, ^{235}U, ^{232}Th, and ^{40}K series and artificial radionuclides such as ^{137}Cs, ^{134}Cs, and ^{90}Sr coming from nuclear fallout following atmospheric nuclear weapon tests and nuclear reactor accidents. Radium isotopes (^{226}Ra and ^{228}Ra), which are found in the ^{238}U and ^{232}Th decay chains, respectively, are the most radiotoxic and dangerous elements. Indeed,

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these elements can quickly enter aquifers due to their high solubility in water and are fixed on the bones after their ingestion because of their similarity with calcium (Sánchez et al., 1999). Once these radionuclides are inside the human body, they will be distributed to all vital organs via the bloodstream as they are very miscible and behave like other non-radioactive isotopes metabolized by the body. Many studies in the world, relating to the identification and quantification of radionuclides with different methods according to their advantages and disadvantages, have been carried out. These techniques can be used for other purposes in the exposure control procedure. Among these techniques are alpha spectrometry, liquid scintillation, mass spectrometry, or gamma spectrometry analysis. This later was used in different countries, including Nigeria (Agbalagba et al., 2013), Jordan (Al-Amir et al., 2012), Germany (Beyermann et al., 2010), Italy (Desideri et al., 2007), Saoudi Arabia (Salih et al., 2002), Greece (Kehagia et al., 2007), USA (Landsberger & George, 2013), Sudan (Alfatih et al., 2008), and Turkey [(Yalcin et al., 2012), (Gorur & Camgoz, 2014)], In Morocco, an effort is being done to identify radionuclides that occur in water, such as the work done by (Hakam et al., 2001), who tested the radioactivity present in spring water in Morocco using the most extensively used chemical precipitation procedure. Their purpose was to evaluate spring mineral waters right before bottling. We extensively employ the gamma spectrometry method in this study to determine radioactive quantities in commercially bottled water in Morocco. The concentration of radioisotopes in bottled mineral water can cause long-term cancers, for example, cancer of the bones and sinuses of the head, by the accumulation of high doses of radium in the bone tissues. Thus, this work aims to establish a reference for natural radioactivity in bottled water in Morocco. To evaluate the average doses received by the inhabitants and compare them to those advised by international organizations, the activity concentration of ^{40}K is calculated. This allows us to determine the radium equivalent index (Ra(eq)), the equivalent annual dose (AED), and the cancer risk coefficient (ELCR), which are very important to prevent radiology risks that may cause damage to public health. Based on the recommendations from the World Health Organization fixing the amount of water that a baby, a child, or an adult can drink annual dose calculations have been performed. In this respect, the WHO has determined that the maximum annual effective reference dose from drinking water consumption is 0.1 $mSv.year^{-1}$ (Graham, 1998). The latest recommendations presented by the WHO for the quality of drinking water from 2011 (WHO, 2011) in radiological aspects request an activity concentration for a wide range of radionuclides of 0.1 $Bq.L^{-1}$ for ^{228}Ra and 1 $Bq.L^{-1}$ for ^{226}Ra. On the other hand, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) has estimated that exposure to natural radionuclides accounts for about 70% of the radiation dose. The global average human exposure from natural sources is 2.4 $mSv.year^{-1}$ and the injection dose is approximately 0.29 $mSv.year^{-1}$ (UNSCEAR, 2000). The data provided in this paper helps to determine the baseline levels of natural radioactivity in drinking water and to collaborate in the formulation of future guidelines for the radiological protection of the Moroccan population.

Materials and methods

Materials

All the experimental data were measured using the equipment of the LPMR laboratory, in particular, the NaI(Tl) scintillator SCIONIX 2"×2" model, coupled to an MCA CAEN multichannel analyzer. All analyses were carried out in the laboratory which is located in the basement. In order to minimize background noise as much as possible, a 5-cm-thick lead shield was used (see Fig. 1).

Methods

Sampling

In this work, twelve bottles of drinking water were randomly purchased from different outlets. These bottles were in good storage condition along with the production and expiration dates. They were well stored in our laboratory away from radioactive or any other radiation sources. The samples were packed to the brim in new 1.20-L Marinelli containers (Fig. 2). Data acquisition duration is set to 24 h, in a way to produce a good spectrum under the same climatic conditions. Indeed, the region of eastern Morocco is known by a large

Fig. 1 A view of setup gamma-ray spectrometry (Right) and sources used for calibration (Left)

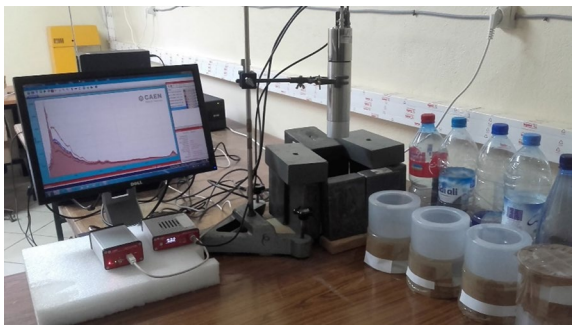

Fig. 2 Samples of drinking water

temperature gradient between day and night which can vary up to 15 °C.

Measurements

Using the trace of radioactive elements present in a sample as shown in Table 1, *P* is the gamma emission probability, and the activity of the parent elements is determined. For example, to determine the activity concentration of ^{226}Ra and ^{228}Ra respectively, the gamma peaks used are 609.01 keV of ^{214}Bi and 583.6 keV of ^{208}Tl. The activity concentration of ^{40}K was measured directly by their gamma line at 1460.8 keV.

The energy calibration, as well as the efficiency calibration "efficiency=f(energy)," was performed using known gamma energy emission sources (^{60}Co, ^{137}Cs, ^{22}Na, …); this stage is included in our previously published work (Bazza et al., 2019). The efficiency of each energy in the resultant spectrum may be determined, applying the formula:

$$Eff = \frac{N}{A(t).p.t}, \tag{1}$$

Table 1 The radioactive elements detected by their daughters using gamma-ray spectrometry

Element	Daughter	Daughter detected	Energy (keV)	*P* (%)
^{238}U	^{226}Ra	^{214}Pb	351.93	37.6
		^{214}Bi	609.01	46.1
			1124	15.1
			1769	15.4
^{232}Th	^{228}Ra	^{228}Ac	911.2	25.8
			973	15.8
			475.65	4.4
		^{212}Pb	238.6	43.3
		^{208}Tl	583.19	84.5
			2661	99
		^{212}Bi	727.33	6.6
^{40}K	^{40}K		1460.8	10.72

with *Eff* which is the efficiency of the photopeak, *N* the net peak area at gamma-ray energy, *A*(*t*) the theoretical activity for the sources used, *p* the emission probability of the gamma-ray, and *t* the live counting time(s).

The activity concentration was calculated by the following equation:

$$A(BqL-1) = \frac{N - Nb}{\varepsilon.p.t.v}, \tag{2}$$

where *N* is the net area at γ-ray energy in the sample spectrum, *Nb* is the net peak area in the background spectrum, ε is the efficiency of the detection, and *v* is the sample volume (L).

The standard MDA (the minimum detectable activity) estimation procedure is based on the equation proposed by Currie in 1968 (Curie, 1968).

$$MDA\left(Bq.L^{-1}\right) = LD.\varepsilon.p.t.v, \tag{3}$$

$$LD = 2.71 + 4.65\sqrt{B}, \tag{4}$$

where LD is the lower limit of detection and B is the number of counts under the net area (background spectrum).

The MDAs of 2^{26}Ra, ^{228}Ra, and ^{40}K calculated for each sample are represented in Table 2.

The MDA should be less than the measured element activity.

Table 2 MDA and mean of each sample

MDA (mBq.L^{-1})	^{226}Ra	^{228}Ra	^{40}K
Min	0.6467	0.1893	8.326
Max	0.7327	0.8259	11.451
Mean	0.5431	0.6870	10.332

Results and discussion

Based on the energy calibration and appropriate regions of interest, the characteristics of each peak in the environmental spectrum are identified. Then, the recognition of radionuclide peaks is carried out, as well as the main dangerous and most toxic radioactive elements which are ^{226}Ra and ^{228}Ra. After calibration and identification of the radionuclides, the activity concentration of the main radioelements in the 12 samples studied in this work was calculated. The factor used to compare the radiological effects of no uniform distribution of ^{40}K, ^{226}Ra, and ^{232}Th nuclei is the Ra(eq), calculated with Eq. 5 given by (Beretka and Matthew, 1985).

$$Ra(eq) = A_{\mathrm{Ra}} + 1.43A_{\mathrm{Th}} + 0.077A_{\mathrm{K}}, \tag{5}$$

Figure 3 shows the activity concentration of ^{226}Ra, ^{238}Th, and ^{40}K for every sample in this study and the radium equivalent value.

From the results obtained in Table 3, it can be seen that the activity concentration of ^{238}U, the maximum is 5.97 mBq.L^{-1} and the minimum is 2.37 mBq.L^{-1}

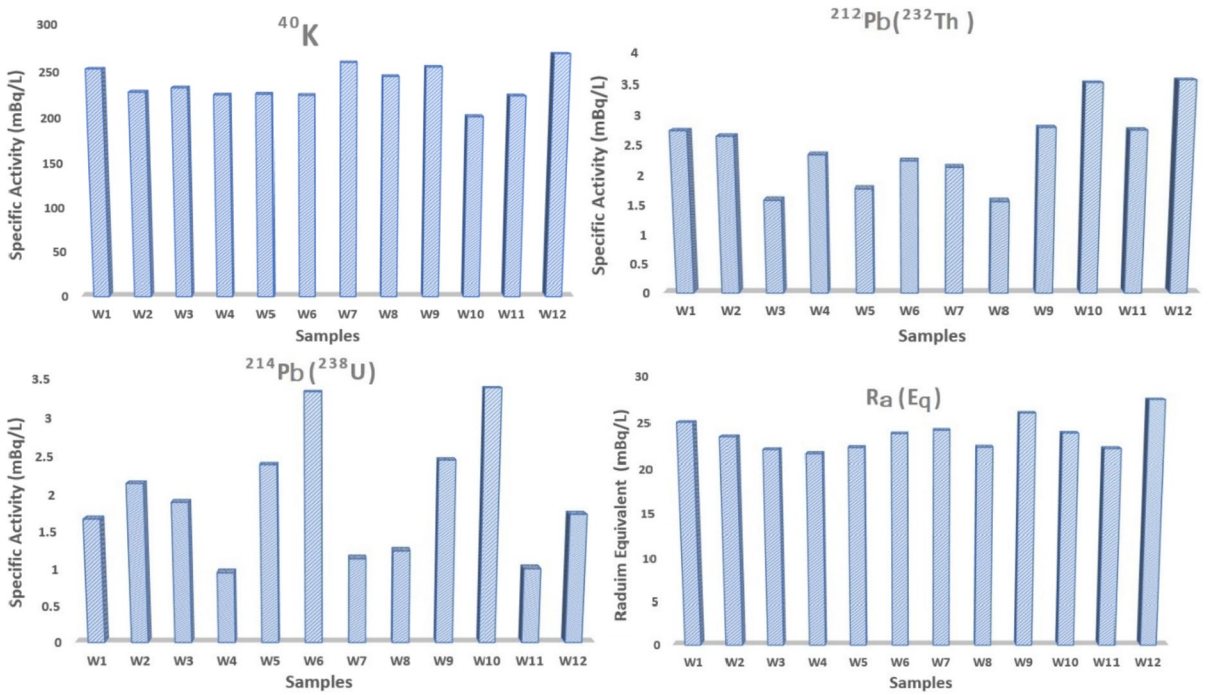

Fig. 3 Specific activity of ^{214}Pb (^{238}U), ^{212}Pb (^{232}Th), and ^{40}K, respectively, and the values of Ra(eq) for each simple

Table 3 Results of activity concentration for ^{40}K, ^{238}U, and ^{232}Th and radium equivalent ($mBq.L^{-1}$) of 12 commercialized water samples

Sample code	Isotope and its identifier			Ra(eq) ($mBq.L^{-1}$)
	^{238}U and ^{226}Ra ^{214}Pb ($mBq.L^{-1}$)	^{232}Th and ^{228}Ra ^{212}Pb ($mBq.L^{-1}$)	^{40}K ($mBq.L^{-1}$)	
W1	1. 674	2.739	252.77	25.03
W2	2.146	2.648	227.62	23.44
W3	1.897	1.575	232.29	22.02
W4	0.951	2.339	224.54	21.57
W5	2.392	1.767	225.43	22.26
W6	3.335	2.238	224.11	23.77
W7	1.142	2.130	259.67	24.18
W8	1.247	1.554	244.60	22.29
W9	2.453	2.792	254.89	26.06
W10	3.386	3.512	200.68	23.85
W11	1.008	2.750	223.51	22.15
W12	1.738	3.560	269.19	27.54
Average	1.947	2.467	236.6	23.68
Maximum value	3.386	3.560	269.19	27.54
Minimum value	0.951	1.554	200.68	21.57
Worldwide [7;26]	35	30	400	370

which are the activity concentrations for sample number 12 and sample number 5 respectively.

For the activity concentration of ^{232}Th, the maximum is 8.74 $mBq.L^{-1}$ and the minimum is 2.28 $mBq.L^{-1}$, and for the activity concentration of ^{40}K, the maximum is 269.19 $mBq.L^{-1}$ and the minimum is 200.68 $mBq.L^{-1}$, which are the activity concentrations for sample number 12 and sample number 10, respectively, with an average of 3.91 $mBq.L^{-1}$ for ^{226}U, 5.63 $mBq.L^{-1}$ for ^{232}Th, and 236.6 $mBq.L^{-1}$ for ^{40}K. The worldwide concentration activity of ^{238}U, ^{232}Th, and ^{40}K is 35, 30, and 400 $Bq.L^{-1}$ respectively (UNSCEAR, 2000).

For the radium equivalent Ra(eq), the maximum activity is 27.54 $mBq.L^{-1}$ and the minimum activity is 21.57 $mBq.L^{-1}$, which are the radium equivalent activity for sample number 12 and sample number 5, respectively, with an average of 23.68 $mBq.L^{-1}$. These levels do not exceed the dose limit (370 $Bq.L^{-1}$) set by the Organization of Economic Cooperation and Development. Making a comparison of activity concentrations of the main radionuclides which are ^{238}U, ^{232}Th, and ^{40}K in the mineral waters of European countries, it could be seen that Moroccan mineral waters are placed in minimum levels with regard to activity concentrations.

The results of studies carried out on mineral waters in certain European countries show a great divergence. Indeed, the concentration of ^{238}U in mineral waters in Slovenia is 0.14 $mBq.L^{-1}$, while in Spain, it is 4000 $mBq.L-^{1}$. On the other hand, ^{232}Th was detected in a small number of analyzed water and its concentration varied in the range of 7.1 to 190 $mBq.L^{-1}$. Furthermore, the concentration of ^{40}K was from 15.7 to 10,064 $mBq.L^{-1}$ (Janković et al., 2012).

In Algeria, the concentration averages of ^{238}U, ^{232}Th, and ^{40}K are 26 $mBq.L^{-1}$, 30 $mBq.L^{-1}$, and 1000 $mBq.L^{-1}$ respectively (Charles, 2001). In Table 4, several works in different countries have been cited to make a comparison to the concentration of the obtained activity.

The radioactivity of all samples was found to be below the 190 values given by the (UNSCEAR, 2000).

Radiation dose estimation

The effective dose arising from the ingestion of a radioisotope can be estimated using the dose coefficient. Data for age-related dose coefficients for ingestion of radionuclides have been published by the ICRP and the International Atomic Energy

Table 4 Comparison of activity concentration of radionuclides in different countries

Origin	Activity concentrations of radionuclides ($mBq.L^{-1}$)		
	^{238}U	^{232}Th	^{40}K
Poland (Benedik & Jeran, 2012)	0.8–437		15.7–10.064
Croatia (Chau & Michalec, 2009)	300–600		
Slovenia (Tanaskovic et al., 2011)	0.14–32		
Serbia (Karamanis et al., 2007)	10–530		200–1130
Greece (Kovacs et al., 2004)	0.6–22.1		
Hungary (Wallner & Jabbar, 2010)	4.3–910		
Austria (Charles, 2001)	2–211		
Brazil (Gans, 1985)	27		
France (Ajayi & Owolabi, 2008)	7–700		
Germany (Beyermann et al., 2010)	0.8–350		
Italy (Ajayi & Owolabi, 2008)	0.2–1200		
Portugal (Gans, 1985)	26.7		
Spain (Ajayi & Owolabi, 2008)	20–4000		
Argentina (Gans, 1985)	4.4		
Algeria (Gans, 1985)	26	30	1000
Hong Kong (Gans, 1985)		7.1	110
Bangladesh (Gans, 1985)		190	4160
Tunisia (Ajayi & Owolabi, 2008)			< 3510
USA (Ajayi & Owolabi, 2008)	0.4–1.8		80–169
Turkey (Ajayi & Owolabi, 2008)	11–36		
Pakistan (Ajayi & Owolabi, 2008)			31–140
Serbia (Janković et al., 2012)	< 70	< 50	< 250
Saudi Arabia (Al-zahrani, 2016)	0.105–0.568	0.016–0.382	18.84
Malaysia (Almayahi et al., 2012)	0.70–7.03	0.55–8.64	22–53
Egypt (El-Gamal & El-Mageed, 2014)	0.07–0.54	0.05–0.5	3.25–8.72
Morocco (present work)	0.95–3.38	1.55–3.56	200.7–269.2

Agency (W. H. Organization, 1996). The natural radionuclides ^{238}U, ^{232}Th, and ^{40}K are responsible for the dose that the organism receives by ingestion mode, that is, through foods and drinking water. The effective annual dose may be calculated using the data from the UNSCEAR (2008) and the formula below:

$$AED = Cr.\sum_{i} DCF_{i}.A_{i}.f_{i} \ , \tag{6}$$

where Cr is the consumption rate intake of norms in water, DCF_i is the dose convection factor for ingestion, A_i is the concentration of the activity in the water sample, and f_i is the gut transfer factor.

The annual consumption rates are 250, 350, and 730 $L.year^{-1}$ for infants, children, and adults, respectively (WHO, 2011). The dose conversion factors for adults given by the (ICRP, 1996) (James & Birchall, 1995) are 2.8×10^{-4}, 6.9×10^{-4}, and 6.2×10^{-4} $mSv.Bq^{-1}$ for ^{238}U, ^{232}Th, and ^{40}K, respectively.

From Table 5 and Fig. 4, it could be seen that sample 12 has the highest radioactivity of all the different ages studied while samples 3 and 5 have the lowest radioactivity. This study confirms that infants are more vulnerable due to bone growth. The average annual effective dose is higher for infants.

The results reported in Table 6 contain the maximum, minimum, and average of the annual doses absorbed by the three different ages. Unfortunately, they show that the obtained values are too far from the doses limited by international institutes and foundations: 0.26, 0.2, and 0.1 $mSv.year^{-1}$ for infants, children, and adults, respectively (WHO, 2011; W. H. Organization, 1996; UNSCEAR, 2000).

Table 5 Value of total annual effective doses for the different samples

Sample code	AED μSv.year^{-1}		
	Infants	Children	Adults
W1	1.4376092	0.984429217	0.78574383
W2	1.32528323	0.91768806	0.73978681
W3	1.2078388	0.783826614	0.5953371
W4	1.25861194	0.856085733	0.67911018
W5	1.20899928	0.798540808	0.6169988
W6	1.2740186	0.868259175	0.6905899
W7	1.38417434	0.914287588	0.70604394
W8	1.24947221	0.80323791	0.60424995
W9	1.46308619	1.005288453	0.8049903
W10	1.33431092	0.981741491	0.8319279
W11	1.30631438	0.908697735	0.73512962
W12	1.61082276	1.133096379	0.92550365

Table 6 Minimum, maximum, and average annual doses absorbed by infants, children, and adults in Morocco

Age	Annual effective doses (μSv.year^{-1})		
	AED_{min}	AED_{max}	$AED_{average}$
Infant (1–6)	1.207	1.610	1.337
Children (7–12)	0.783	1.133	0.912
Adult > 17	0.595	0.925	0.725

Table 7 The maximum and the average excess lifetime cancer risk (ELCR) for infants, children, and adults

	ELCR maximum	ELCR average
Infants	5.63×10^{-6}	4.68×10^{-6}
Children	3.95×10^{-6}	3.19×10^{-6}
Adults	3.25×10^{-6}	2.53×10^{-6}

Excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR) was calculated using Eq, (7) (UNSCEAR, 2000):

$$ELCR = AED \times DL \times RF, \quad (7)$$

where AED is the annual equivalent dose Sv.year^{-1}, DL is the average duration of life (estimated to be 70 years), and RF is the risk factor. The ICRP uses RF as 0.05 Sv^{-1} for public (UNSCEAR, 2000).

The maximum, the mean of the AED of all the samples studied, and Eq. 5 were used to calculate the ELCR. Thus, the obtained values will be compared to the international norms and limits given by the ICRP (James & Birchall, 1995).

In Table 7, the ELCR of the maximum and the average are 5.63×10^{-6} and 4.68×10^{-6}, respectively; they are lower than 2.5×10^{-3}, the maximal value proposed by (James) . As we see, infants are more sensible because of their organ composition. Figure 5

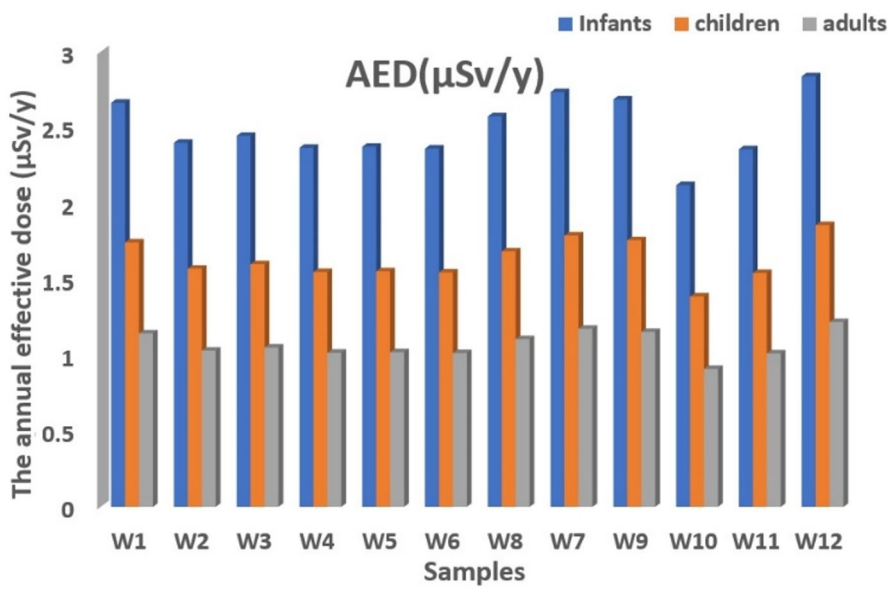

Fig. 4 Annual doses absorbed by infants, children, and adults in Morocco

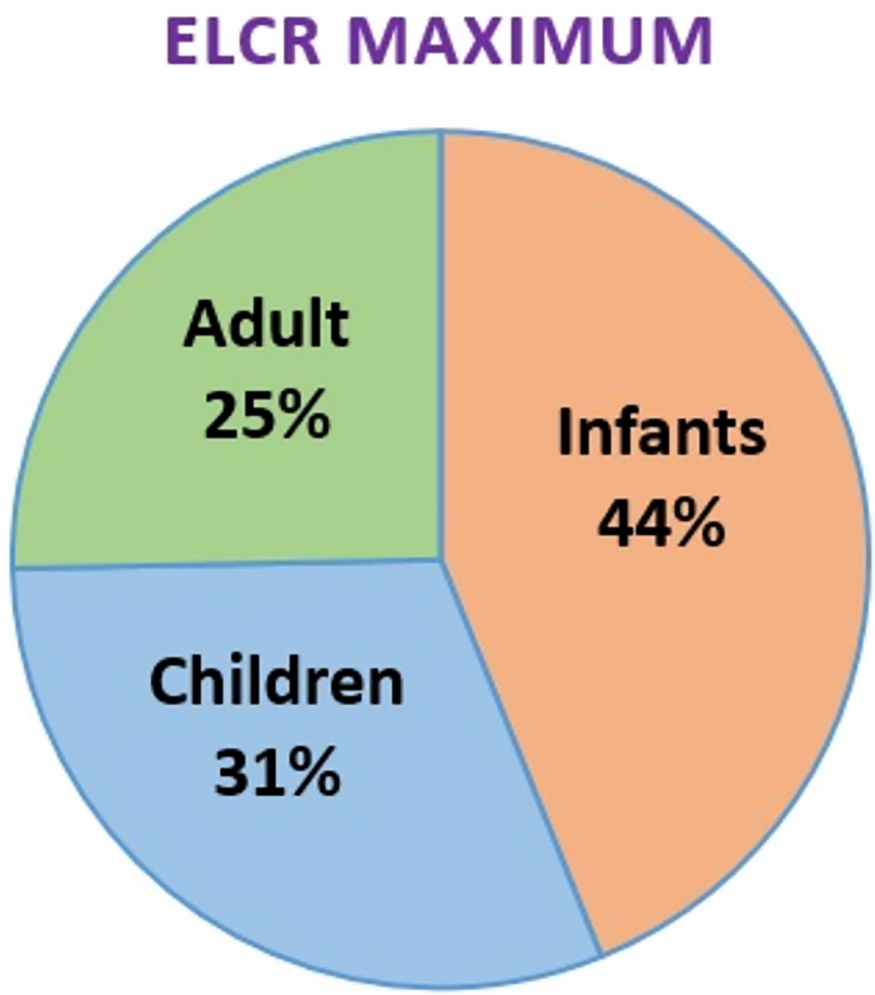

Fig. 5 Excess lifetime cancer risk (ELCR) for different ages in Morocco

shows the percentage of cancer risk for Infants, children, and adult.

Conclusion

Radionuclides are toxic and dangerous and can provoke serious damage to human health. The need to assess the concentration of radioactive elements in bottled water is necessary to reassure consumers. In this vision, the present study, which is a pioneer in Morocco, intends to evaluate the natural radioactivity present in mineral water bottles by using gamma spectrometry. The measurement of the activity concentration of ^{238}U, ^{232}Th, and ^{40}K in 12 samples of mineral water bottled in Morocco allowed us to calculate indices, such as the radium equivalent (Ra(eq)), the annual effective dose absorbed by the public, and the excess lifetime cancer risk (ELCR). The obtained data are compared to the reference values proposed by the committees and the national and the international organizations. The results could be used as a reference base to guide local and global research in radiation protection. All indices calculated in this study were below the recommended indicative levels. The maximum value measured of the radium equivalent Ra(eq) was 27.54 mBq.L^{-1} which is very lower than 370 Bq.L^{-1}, the limit activity fixed by (OECD). Furthermore, the maximum effective annual dose (AED) measured in infants is 1.61 μSv.year^{-1}. It is also very small than 29 μSv.year^{-1}, the value which represents 10% of 290 μSv.year^{-1}, the dose recommended by the

UNSCEAR for the ingestion mode of natural ionizing radiation. Finally, the ELCR cancer development risk index value was 5.63×10^{-6}, while the maximum value proposed by the ICRP is 2.5×10^{-3}.

It could be concluded that the mineral drinking water bottled in Morocco does not exhibit any radiological peril to the health of consumers. This prompts us to carry out analyses, in future works, on other samples of the most common foods in this region to predict the total value of the annual dose absorbed by the population due to food.

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