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ORIGINAL ARTICLE

Natural radioactivity levels and evaluation of radiological hazards from Beni Haroun dam sediment samples, northeast Algeria

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Abstract Levels of naturally occurring radioactivity in sediment samples of Beni Haroun dam have been investigated. The activity concentrations of ^{238}U and ^{232}Th decay chains and ^{40}K primordial radionuclide have been measured using high-resolution HPGe detector. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides were found in the ranges 9–66, 14–37, and 177–288 Bq/kg with the mean values 24.67, 25.98, and 208.10 Bq/kg, respectively. Radiological hazard parameters were estimated based on the activity concentrations for ^{226}Ra, ^{232}Th, and ^{40}K to find out any radiation hazard associated with the sediments. Correlation studies between pairs of radionuclides were performed and discussed, and the obtained results are compared with international recommended values.

Keywords Beni Haroun · Gamma spectrometry · Radionuclides activity concentration · Radiological parameters

Introduction

Terrestrial gamma radiation in the sediment is mostly coming from decays of uranium (^{238}U) and thorium (^{232}Th) series, together with a primordial radionuclide ^{40}K. The levels of natural activity are related to the types of rock or soil from which the sediment originates (UNSCEAR 1982; UNSCEAR 2000; Ravisankar et al. 2015; Krmar et al. 2013). These radionuclides can be transported by various aqueous systems like the rivers from short to long distances. They can be sequestered by diverse geological materials and finally collected from the bottom of dam lakes or deposited in marine sediments (UNSCEAR 2000). Man is always exposed to the natural radiation originating from the soil, since the main path for the transfer of radionuclides to human body is the soil–plant–man chain. Hence, the studies of natural radioactivity allow the assessment of the radiological risk to the human health and to provide useful information in the monitoring of environmental radioactivity. Many authors have studied the natural background radiation levels in different environmental samples (Sam et al. 1997; Yii et al. 2009; Narayana et al. 2007; Morsy et al. 2012; Ugur et al. 2012; Saç et al. 2012; Ergül et al. 2013; Krmar et al. 2013; Sivakumar 2014; Sankaran Pillai et al. 2015). Beni Haroun dam was chosen as a subject of our investigation. It is one of the biggest dams in Africa and the largest one in Algeria. Lake of the dam (36°33′ 19″ N and 6°16′11″ E) is located in the northeast of Algeria (Fig. 1a), at about 40 km north of Constantine city. The dam was built on the confluence of Wadi Rhumel (crossing Constantine city) and Wadi Endja (crossing Mila town) which are main tributaries of Wadi El Kebir. It covers an area of 3.929 ha, and it is 120 m in depth with a capacity of 960 million cubic meters of water. The climate of Beni Haroun basin is changing from semiarid in the south to humid and rainy in the north (Mebarki et al. 2008, El-Hadef El-Okki et al. 2013). This dam supplies several areas by pure water in the northeast part of the country. Also, it provides a significant amount of irrigation water for a few hundred hectares of farms in neighboring regions. The aim of this study was to evaluate

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Fig. 1 **a** Beni Haroun dam location, **b** sampling location throughout Beni Haroun dam

the natural radioactivity levels in sediment samples collected from different locations of Beni Haroun Lake and around. The measurements of the activity concentration of ^{238}U, ^{232}Th, and ^{40}K in sediment samples were carried out using gamma-ray spectrometry in a low background configuration. Results of this analysis obtained for ten samples collected from the dam are presented in this paper. An assessment of the radiological risks on humans due to the natural radioactivity arising from sediment samples was determined in terms of the radiological parameters (Ra_{eq}, H_{ex}, H_{in}), absorbed dose (ADR), and annual effective dose (AED).

Materials and methods

Sampling and sample preparation

Ten surface sediment samples with a depth of 0–10 cm were collected from several different locations along the dam and nearby as it displayed in Table 1, between June and October 2014 to determine the natural radionuclides distribution in the surveyed area, as it is shown in Fig. 1b. In the laboratory, each sediment sample was heated at 80 °C for 48 h in order to remove the moisture until a constant weight was reached. Then it was sieved to fine powder, passed through a standard 1-mm mesh size and sealed into small cylindrical plastic containers approximately during 30 days ($\sim$ 7 half-lives) to reach a secular equilibrium between the ^{238}U and ^{232}Th series and their respective progeny before measurement (Santawamaitre et al. 2014; Bajoga et al. 2015).

Measurements

The ^{238}U and ^{232}Th decay chains and ^{40}K activity concentrations were determined by the gamma-ray spectrometry technique using a coaxial hyper-pure germanium (HPGe) detector. This later was enclosed inside by 11.4-cm-thick graded lead shield (Canberra 747 Series Lead Shield) with liners of 3 mm of tin and 0.15 cm of copper to reduce X-ray fluorescence from lead shield. The gamma-spectrometry system has a measured gamma-energy photopeak resolution of 1.85 keV at 1332.5 for ^{60}Co and 0.86 keV at 122 keV for ^{57}Co. The spectrum treatment was carried out using the Genie 2000 software. The detector

Table 1 Sampling locations

Sample sampling point *e*	Geographical position		
	Locations	Latitude (N°)	Longitude (E°)
S1	Siliana	36° 32′ 40.29″	6° 17′ 28.15″
S2	The dam dike	36° 34′ 01.96″	6° 16′ 43.59″
S3	Wadi Mila	36° 29′ 42.14″	6° 17′ 43.86″
S4	Sidi Meroun	36° 29′ 57.09″	6° 24′ 33.90″
S5	Pont G-M S1	36° 32′ 40.29″	6° 18′ 30.23″
S6	Pont G-M S2	36° 30′ 40.96″	6° 18′ 43.83″
S7	Wadi Ouerzeg	36° 29′ 12.76″	6° 25′ 35.85″
S8	Wadi Metlili	36° 30′ 27.45″	6° 30′ 40.96″
S9	Downstream the dam	36° 34′ 06.4″	6° 16′ 21.57″
S10	Aress (Boughardeine)	36° 29′ 30.81″	6.06° 46′ 11″

efficiency calibration was measured using ^{152}Eu liquid source mixed in sediment matrix. The coincidence summing effects on efficiency calibration were taken into account to correct the efficiency values by the Monte Carlo simulation (Azbouche et al. 2015). The counting geometries for the source and sediment samples were identical. The measured full-energy peak efficiency data were fitted to the response function in the form:

$$\varepsilon_\gamma = y_0 + A_1 e^{-\frac{E_1}{t_1}} + A_2 e^{-\frac{E_2}{t_2}} \quad (1)$$

where y_0, A_1, and t_1 and t_2 are the fitting function parameters, and ε_γ the efficiency at energy E. The fitted curve for this full-energy peak absolute response function is shown in Fig. 2.

Activity concentration determination

Each prepared sediment sample was measured during 48 h using the coaxial hyper-pure germanium detector. The ^{238}U activity concentration presenting in the sample was estimated by means of ^{234}Th 63.38 keV gamma-ray energy. The activity of ^{226}Ra was determined from the gamma lines associated with the short-lived daughters of ^{214}Bi 609.14 keV and ^{214}Pb 351.81 keV after the secular equilibrium. ^{232}Th activity was determined by the 911.07, the 510.59, and the 238.56 keV gamma line from ^{228}Ac, ^{208}Tl, and ^{212}Pb, respectively. The gamma-ray peak associated with the decay from ^{40}K at 1460.67 keV was used to determine the activity concentration of this nucleus. We

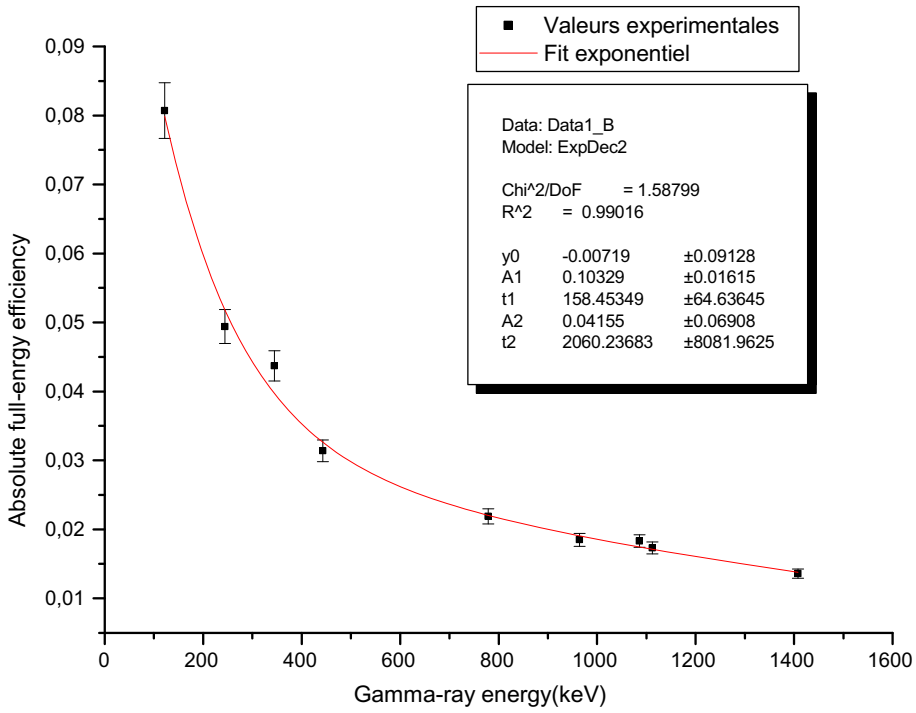

Fig. 2 Efficiency curve of ^{152}Eu source

remind that specific activity of an individual radionuclide in sediment samples is given by the following equation:

$$A_s = \frac{N_E}{\varepsilon_\gamma p_\gamma t_c m} \quad (2)$$

where A_s is the activity concentration of a particular nuclide in the units of Bq/kg, N_E the net count at the interest peak energy (corrected by background counts of the corresponding full-energy peak), ε_γ the absolute full-energy peak detection efficiency, P_γ the gamma-ray emission probability, t_c the counting time in second, and m the mass of sediment sample in kilogram.

The minimum detectable activity (MDA) for each radioelement is given by (G. Gilmore and J.Hemingway 1995):

$$\mathrm{MDA} = \frac{\mathrm{DL}}{\varepsilon_\gamma p_\gamma t_c m} \quad (3)$$

where DL is the detection limit under the background counts B, such that:

$$\mathrm{DL} = 2.71 + 4.65\sqrt{B}$$

Table 2 shows MDA values of radionuclides for investigation sediment samples.

Results and discussion

Radioactivity levels of sediment samples

The specific activities of natural radionuclides measured in sediment samples are presented in Fig. 3. The results show an inhomogeneous distribution of these elements at different sampling locations. This difference is due essentially to the variation in the geological composition of the studied area. In all samples, ^{40}K has the greatest activity compared with other isotopes. This can be explained by the presence of clay, which contains relatively high concentration of potassium. ^{232}Th activity was higher than ^{226}Ra in all sediment samples expected S9 and S10 ones. These elements have different behaviors in the sediments: the ^{238}U (^{226}Ra) dissolves in water; ^{232}Th is a particularly insoluble element, and it is usually found associated with a solid matter. The highest value for ^{226}Ra level observed in sample S10 reflects some influence of the intensive farming activities in this region. But in comparison with the increased value of ^{226}Ra in sample S9, it could be explained by the human activities on the Rhumel and Endja river banks. The results of our present work have been compared with the world mean activity reported by United Nations Scientific Committee on the Effects of Atomic Radiation, as it is shown in Fig. 3. ^{40}K specific activities in all samples are less than the standard average values of 400 Bq/kg (UNSCEAR 2000). Unlike the obtained ^{232}Th values in studied samples, which are lower than the standard average, we noted a slight elevation of the measured activity concentration in sample S9 compared to the standard universal mean of 33 Bq/kg (UNSCEAR 2000). The ^{226}Ra activity concentrations of S9 and S10 samples are higher than the standard average value 35 Bq/kg (UNSCEAR 2000). This increase is a consequence of the reasons mentioned above.

Table 2 MDA values of radionuclides

Radionuclides	E (keV)	MDA (Bq/kg)
^{40}K	1461.67	12.61
^{214}Pb	351.81	0.87
^{214}Bi	609.14	1.14
^{208}Tl	510.59	4.33
^{212}Pb	238.60	0.45
^{228}Ac	911.07	1.58

Correlation studies

Correlation studies were made between each radionuclides pair in order to find extent of these radionuclides together at a particular place. Scatter graphs in Fig. 4 show a very low correlation between (^{226}Ra, ^{40}K) and (^{226}Ra, ^{232}Th) pairs in sediment samples, where the correlation coefficient is $R = 0.025$ and 0.18, respectively. However, there is a good correlation between (^{232}Th, ^{40}K) pair with $R = 0.71$. This strong correlation indicates that the individual result is a good predictor for the concentration of the other component of the pair (Darwish et al. 2013).

Activity ratios

^{232}Th/^{226}Ra, ^{232}Th/^{40}K, and ^{226}Ra/^{40}K concentration ratios were evaluated from the measured values of activity concentrations and are summarized in Table 3. The results illustrate that all the ^{232}Th/^{40}K and ^{226}Ra/^{40}K ratios are lower than 1 which indicate that the ^{40}K activity concentrations are higher than those of ^{226}Ra and ^{232}Th in these samples. Furthermore, most samples have ^{232}Th/^{226}Ra ratios higher than 1 meaning that the sediments are enriched in ^{232}Th more than ^{226}Ra. Conversely for S9 and S10 samples, it can be seen that the ratios are < 1. Thus, the concentration of ^{226}Ra is dominating in comparison with ^{232}Th one in these two samples.

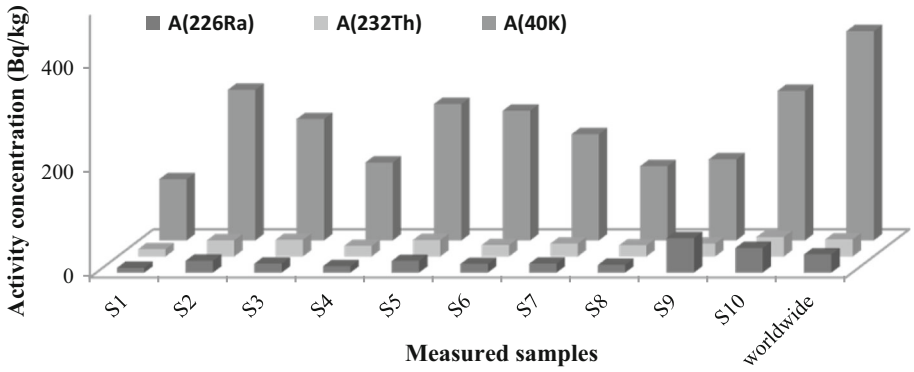

Fig. 3 Activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K for the investigated samples

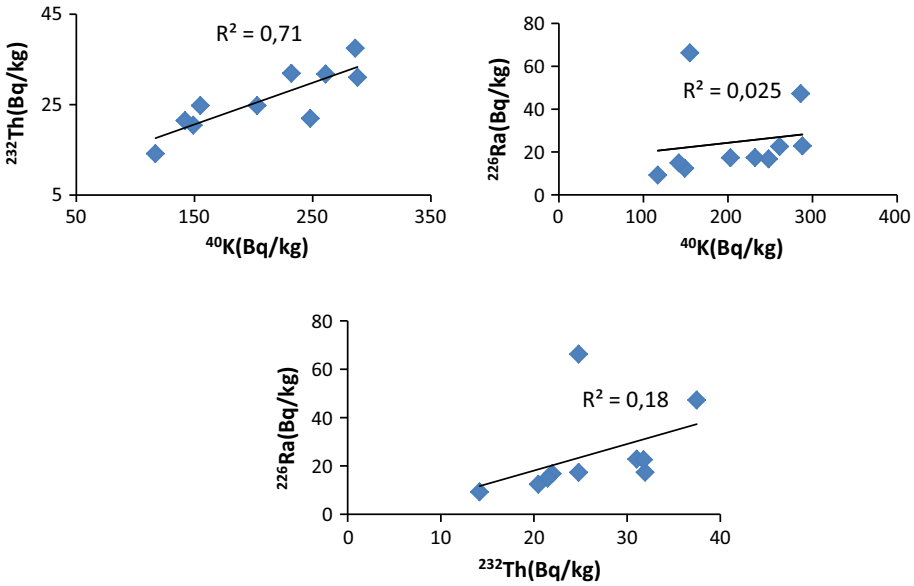

Fig. 4 Correlation between specific activities of natural radionuclides in Beni Haroun dam

Table 3 Activity ratios of ^{226}Ra, ^{232}Th, and ^{40}K

Sample code	Activity ratios		
	^{232}Th/^{226}Ra	^{232}Th/^{40}K	^{226}Ra/^{40}K
S1	1.54	0.12	0.08
S2	1.36	0.11	0.08
S3	1.84	0.14	0.07
S4	1.65	0.14	0.08
S5	1.41	0.12	0.09
S6	1.31	0.09	0.07
S7	1.43	0.12	0.09
S8	1.44	0.15	0.10
S9	0.37	0.16	0.43
S10	0.79	0.13	0.17
Mean	1.31	0.13	0.13

Radiological parameters

Radium equivalent activity (Ra_{eq})

Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{238}U and ^{232}Th decay chains and ^{40}K. It is given by a single quantity (Ra_{eq}), which takes into account the radiation hazards (Beretka and Matthew 1985; Saç et al. 2012):

$$Ra_{eq} = 0.077A_K + 1.43A_{Th} + A_{Ra} \quad (4)$$

where A_k, A_{Th}, and A_{Ra} are the specific activities of ^{40}K, ^{238}Th, and ^{226}Ra in Bq/kg, respectively. It has been assumed that 370 Bq/kg of ^{226}Ra or 259 Bq/kg of ^{232}Th or 4810 Bq/kg of ^{40}K produces the same gamma dose rate. The radium equivalent activity in these sediment samples ranges from 38 to 122 Bq/kg with a mean value of

Table 4 Radiation hazard parameters for sediment samples

Samples	Radiological parameters					
	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	I_γ	D (nGy/h)	AED (μSv/y)
S1	38.48	0.10	0.13	0.28	17.73	21.76
S2	89.35	0.24	0.30	0.65	41.32	50.71
S3	80.87	0.22	0.27	0.59	37.12	45.56
S4	53.15	0.14	0.18	0.39	24.40	29.95
S5	88.06	0.24	0.30	0.64	40.58	49.80
S6	67.23	0.18	0.23	0.50	31.29	38.40
S7	68.40	0.18	0.23	0.50	31.51	38.67
S8	56.54	0.15	0.19	0.41	25.90	31.78
S9	113.60	0.31	0.49	0.79	52.19	64. 04
S10	122.80	0.33	0.46	0.88	56.52	69.36
Range	38–122	0.10–0.33	0.13–0.49	0.28–0.88	17–56	21–69
Mean	77.85	0.21	0.28	0.56	35.86	44
Worldwide	370	1	1	1	59	70

77.85 Bq/kg (Table 4) which is less than the recommended maximum value 370 Bq/kg (Beretka and Matthew 1985). It indicates also that no significant radiological hazard is associated with the sediments.

Gamma representative level index (I_γ)

The representative level index (I_γ) is used to estimate the level of gamma radiation hazards associated with natural gamma emitters in the sediments. It is calculated from the following expression (NEA-OECD 1979).

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (5)$$

It is very important to mention that it must be less than unity in order to keep the radiation hazard insignificant. The calculated values of I_γ vary from 0.28 to 0.88 with a mean value of 0.56 (Table 4). The mean value in the studied area is below than the required which indicates that the sediments do not give rise to any significant radiation hazards.

External and internal hazard indices (H_{ex}, H_{in})

The external hazard index (H_{ex}) is used to assess the radiological suitability of a material (Ravisankar et al. 2015). The important objective of this index is to limit the radiation dose to the admissible dose equivalent limit of 1 $mSvy^{-1}$ (ICRP 60 1990; Hamarneh and Awadallah 2009) and can be obtained as follows:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (6)$$

In addition to the external hazard index, internal exposure to radon and its products are quantified by estimating the internal hazard index (H_{in}). Inhalation of alpha particles emitted from the short-lived radionuclide radon (^{222}Rn), the daughter of the ^{226}Ra (^{238}U), and thoron (^{220}Rn), the daughter product of ^{224}Ra (^{232}Th), is also hazardous to the respiratory organs. This hazard can be quantified by internal hazard index (H_{in}) (Beretka and Matthew 1985; Xinwei 2005) as follows:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (7)$$

The values of H_{ex} and H_{in} must be lower than unity (Diab et al. 2008). The calculated values of these indices for studied samples are presented in Table 4, for different locations. The average values of H_{ex} and H_{in} have been found to be 0.21–0.28, respectively. These results indicate that no elevated radiological health hazards to the people living in nearby terrestrial areas of the sampling sites.

Doses assessment

Absorbed dose rate in air

In order to assess radiological risk, external exposure to radiation arising from naturally occurring radionuclides can be determined in terms of the absorbed dose rate in air at 1 m above the ground surface. The mean activity concentration of ^{238}U, ^{232}Th, and ^{40}K in units of Bq/kg in the sediment samples can be used to calculate the absorbed dose rate using the relation (UNSCEAR 2000):

$$ADR(\text{nGy/h}) = 0.462A_{Ra} + 0.621A_{Th} + 0.041A_K \quad (8)$$

From Table 4, the estimated absorbed dose lies from 17 to 56 nGy/h with a mean value of 35.86 nGy/h. The

obtained results in this work are lower than the standard mean value of 59 nGy/h (UNSCEAR 2000).

Annual effective dose equivalent (AED)

The annual effective dose equivalent for the adults from outdoor gamma radiation is obtained by taking into account the conversion coefficient of 0.7 Sv/Gy from the gamma-absorbed dose rate in air (ADR), and including the outdoor occupancy factor of 0.2 (UNSCEAR 2000). The annual effective dose equivalent in μSv/y resulting from the absorbed dose values (ADR) can be calculated using the following formula:

$$\mathrm{AED}(\mu\mathrm{Sv/y}) = \mathrm{ADR}(\mathrm{nG/h}) \times 365.25 dy^{-1} \times 24 hd^{-1} \times 0.2 \times 0.7 Sv/Gy \times 10^{-3} \quad (9)$$

In Table 4, the calculated annual effective equivalent varies from 21 to 69 μSv/y with a mean value of 44 μSv/y which is lower than the standard average of 70 μSv/y (UNSCEAR 2000). This indicates that the hazardous effects of these radiations are negligible.

Conclusion

In this present study, the natural radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K have been measured in sediment samples collected from different locations inside and around Beni Haroun dam using a gamma-ray spectrometry. The activity concentrations measured across all the samples varied from 9–66, 14–37, and 177–288 Bq/kg with mean values 24.67, 25.98, and 208.10 Bq/kg, respectively. The results show that the distribution of ^{226}Ra, ^{232}Th, and ^{40}K in sediment samples is not uniform which can be explained by a geological structure variation and overuse of fertilizers around the dam. The absorbed dose rate and the annual effective dose equivalent have been evaluated with their radiological risk parameters. Our results can be used as a baseline for the observation of any possible change in the future.

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