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# Measurement of Natural Radioactivity in Some Commercial Marble Samples Used in Algeria

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## Abstract

Ionizing radiations emitted by natural radionuclides have always existed in building materials which directly affect human beings, and can contribute in accumulated dose in long term. The objective of this work was an investigation of natural radioactivity levels associated radiation hazard in commercial marble samples used in Algeria. Six samples were collected from marble dealers and were analyzed by gamma spectrometry, using a high resolution HPGe semi-conductor detector with (1.11 keV for $^{214}$Bi 609 keV line). The spectra were analyzed using the Genie 2000 software dedicated to the processing of gamma spectra. The activity concentrations of primordial radionuclides $^{232}$Th, $^{226}$Ra and $^{40}$K were determined; they ranged from 0.1 to 1.5 Bq/kg with an average concentration value of 0.9 Bq/kg, from 0.5 to 40.3 Bq/kg with a mean concentration value of 9.3 Bq/kg and from 0.7 to 38.7 Bq/kg with an average concentration value of 25.5 Bq/kg, respectively. The results were found to be lower than the world wide average values. In order to assess the radiological hazard associated to these radionuclides, radiation hazard parameters and dose estimations were elaborated.

**Keywords:** Ionizing radiations, Marble, Natural radioactivity, Gamma spectrometry, HPGe detector

## 1. INTRODUCTION

It is quite important to monitor any release of natural radioactivity present in building materials because it makes it possible to asses any chance of radiological hazard to man generated by those materials [1, 2]. The latter are used and dealt with according to standards and guidelines developed as a result of such knowledge [2].

Primordial radionuclides such as the Uranium ($^{238}$U), Thorium ($^{232}$Th) and Actinium ($^{235}$U) series besides Potassium ($^{40}$K) contribute to the major part of the dose rate of natural radioactivity. However, cosmic radiation and cosmogenic radionuclides participate also in this value [1].

There has been an increased interest in studying the radioactivity contained in building materials worldwide [3-6] and many research woks have been conducted in Algeria [7-10]. Marble has gained an infinite popularity as it is widely used in building constructions, mainly as a finishing material. Such fame calls for an inclusive, comprehensive and broad investigation of the effects of this material on the health of the inhabitants.

The main aim of this research wok is to examine the activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in six marble samples using a high-resolution coaxial hyper-pure germanium gamma spectrometry system. The data obtained were used to calculate approximately the absorbed dose rate in the air and the annual effective dose rate. They also helped to determine radiological hazard indices as radium equivalent, external and internal radiation indexes, and representative level index. A comparison between the obtained values in this study and the world wide average ones was made.

## 2. MATERIALS AND METHODS

### 2.1 Samples Collection and Preparation

The six samples of the current study were collected from marble dealers; they were locally extracted from quarries of Constantine (M1), Chelghoum (M2), Guelma (M3), Skikda (M4), Tamanraset (M5) and Tlemcen (M6). The samples were crushed and milled into fine powder, and then they were dried in an oven under a temperature of 110 $^{\circ}$C for at least 24 hours in a way to remove the moisturizing content and obtain a constant weight. The samples were conditioned in thin Plexiglas boxes to avoid the self-absorption of low energy gamma. These boxes were carefully sealed for a period of four weeks (~ 7 half-lives) to ensure secular equilibrium between the $^{238}$U and $^{232}$Th series and their progenies.

### 2.2 Experimental Method for Gamma Spectroscopy

Samples measurements were carried out with a high purity germanium diode of 210 cm$^3$ (n type) detector to determine the gamma spectrum of $^{238}$U, $^{232}$Th decay chains and $^{40}$K. The detector was surrounded by a 30 cm thick archaeological lead shield with a 5 cm copper sealed box for the performance of a nitrogen sweep to eliminate radon contamination, and 20 cm of ordinary lead (low radioactivity less than 30 Bq / kg). The gamma-energy photo-peak resolution of the gamma spectrometry system is 1.11 keV at 609 keV for $^{214}$Bi and 0.95 keV at 352 keV for $^{214}$Pb. The detector efficiency calibration was performed using $^{152}$Eu and $^{232}$Th sources in order to calibrate high energies up to 2614 keV. The Genie 2000 software was used for the acquisition and treatment of the collected data. Each sample was measured for at least 24 hours to obtain the $\gamma$-spectrum with good statistics. The detector efficiency values were calculated by Monte Carlo simulation using Geant3 code. Figure 1 shows the calculated efficiency curves of the detector as a function of gamma rays energy. The gamma-ray transitions of energies 351.9 keV ($^{214}$Pb) and 609.3 keV ($^{214}$Bi) were used to determine the activity concentration of $^{226}$Ra, the gamma-ray transitions of energies 338.4 keV ($^{228}$Ac), 583.3 keV ($^{208}$Tl), 2614 keV ($^{208}$Tl) and 911.1 keV ($^{228}$Ac) were used to determine the activity concentration of the $^{232}$Th and the 1460 keV gamma-ray transition of $^{40}$K was used to determine its concentration in different samples.

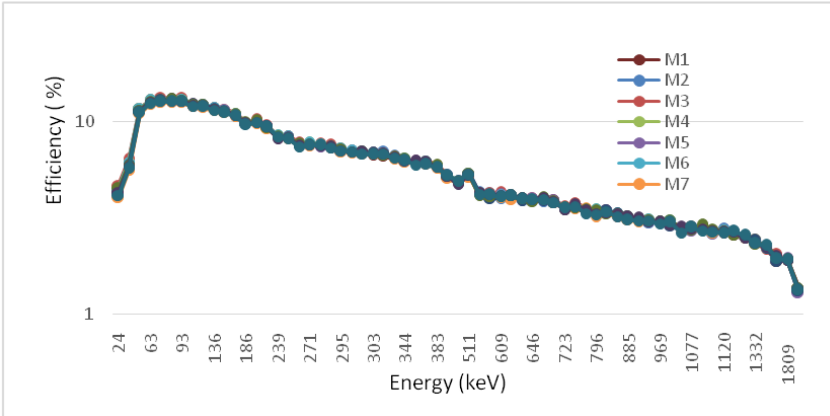



FIG. 1. Efficiency curve as a function of energies.

## 3. RESULTS AND DISCUSSION

### 3.1 Radioactivity Levels of Marble Samples

The activity concentrations of the different marble samples are presented in Figure 2. The results show heterogeneity of the distribution of radionuclides in the samples. This can be attributed to the variation in the geological composition of each region and to the mineral content of the marble. The activity concentrations of $^{232}$Th, $^{226}$Ra and $^{40}$K range from 0.1 to 1.5 Bq/kg with an average concentration value of 0.9 Bq/kg, from 0.5 to 40.3 Bq/kg with a mean concentration value of 9.3 Bq/kg and from 0.7 to 38.7 Bq/kg with an average concentration value of 25.5 Bq/kg, respectively. The mean concentration values are lower than the world wide average concentrations which are 40, 40 and 400 Bq/kg respectively, as it is reported in the European recommendations based on a literature study [11,12]. It is obvious from the results displayed in Figure 2 that the sample M5 contains the highest activity concentrations of different radionuclides. It is noticed that there is an absence of $^{232}$Th and $^{40}$K in the samples M1, M3 and M4. We also observe the absence of artificial radionuclides in the studied samples. This induces that the sampling sites do not present any contamination.

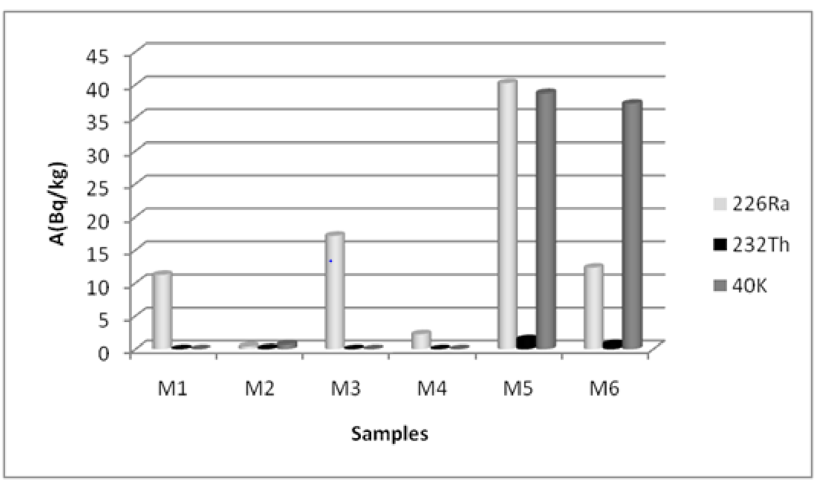



FIG. 2. Activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K for the investigated samples.

### 3.2 Doses Assessment

The absorbed dose rate in the air at 1m above the ground surface which assesses the external exposure to the arising radiation from natural radionuclides and the annual effective dose equivalent that estimates the dose received by adults from outdoor gamma radiation exposure can be determined using relations 1 and 2 [13].

$$D(nGy.h^{-1}) = 0.462A_{Ra} + 0.621A_{Th} + 0.041A_K \quad (1)$$

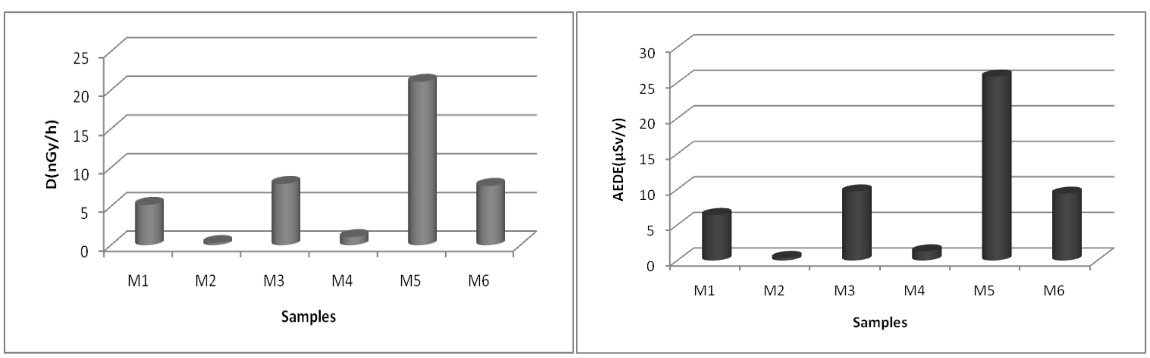
$$AEDE(\mu Sv.h^{-1}) = D(nGyh^{-1}) \times 8760h \times 0.2 \times 0.7SvGy^{-1} \times 10^{-3} \quad (2)$$

Where $A_{Ra}$, $A_{Th}$ and $A_K$ are activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ respectively. The results of calculated absorbed dose rates are shown in Figure 3 (a). They range from 0.30±0.02 nGy/h for M2 to 21.1±0.18 nGy/h for M5, with an average value of 5.3±0.08 nGy/h. This is lower than the recommended safety limit value of 59 nGy/h [15]. The calculated values of annual effective dose equivalent are presented in Figure 3 (b), they vary from 0.37±0.03 μSv/y for M2 to 25.87±0.22 μSv/y for M5 with a mean value of 6.5±0.1 μSv/y which is below the recommended maximum value 70 μSv/y [13].

### 3.3 Radiological Hazard Parameters Determination

The distributions of $^{226}Ra$, $^{232}Th$ and $^{40}K$ are not uniform in the samples. Hence, Radium equivalent ($Ra_{eq}$) has been introduced to estimate the actual activity level of these radionuclides and to assess the radiation hazards associated with their presence in building materials [10,14]. It is defined by equation 3 [3,15].

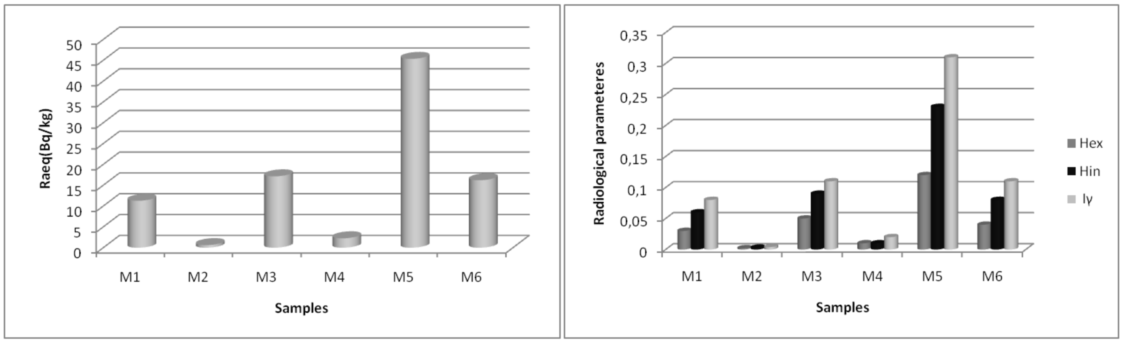
$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3)$$




(a) (b)

FIG.3. (a) Absorbed dose rate in air and (b) Annual effective dose equivalent for the studied samples.

The base estimation of this formula is that 1 Bq/kg of $^{226}Ra$, 0.7 Bq/kg of $^{232}Th$ and 13 Bq/kg of $^{40}K$ produce the same γ-ray dose rate. The $Ra_{eq}$ calculated values are shown in Figure 4 (a), they range from 0.65±0.05 Bq/kg for M2 to 45.34±0.39 Bq/kg for M5, with a mean value of 11.39±0.18 Bq/kg. This latter is below the permissible limit of 370 Bq/kg [15].




(a) (b)

FIG. 4. (a) Radium equivalent and (b) Radiological parameters for the six samples.

The external hazard index ($H_{ex}$) is used to evaluate external γ radiation doses; it is derived from the relation of $Ra_{eq}$. The internal hazard index ($H_{in}$) was introduced to quantify the internal exposure to radon and its short lived progenies. In addition, the level of gamma radiation hazard associated with the natural radionuclides in building materials are estimated using the representative level index ($I_{\gamma}$). They are defined by relations 4, 5 and 6 [3,15].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{258} + \frac{A_{K}}{4810} \quad (4)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \quad (5)$$

$$I_{\gamma} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500} \quad (6)$$

As can be perceived from Figure 4.(b), the calculated values of $H_{ex}$, $H_{in}$ and $I_{\gamma}$ vary from 0.002±0.000 to 0.12±0.00 with a mean value of 0.03±0.00, from 0.003±0.006 to 0.23±0.02 with an average value of 0.06±0.01 and from 0.004±0.000 to 0.31± 0.00 with a mean value of 0.08±0.00, respectively. The resulted data in this study are less than the recommended value (unity).

## 4. ACKNOWLEDGEMENTS

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