Natural radionuclide and radiological assessment of building materials in high background radiation areas of Ramsar, Iran

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ABSTRACT

Building materials, collected from different sites in Ramsar, a northern coastal city in Iran, were analyzed for their natural radionuclide contents. The measurements were carried out using a high resolution high purity Germanium (HPGe) gamma-ray spectrometer system. The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K content varied from below the minimum detection limit up to 86,400 Bqkg^{-1}, 187 Bqkg^{-1}, and 1350 Bqkg^{-1}, respectively. The radiological hazards incurred from the use of these building materials were estimated through various radiation hazard indices. The result of this survey shows that values obtained for some samples are more than the internationally accepted maximum limits and as such, the use of them as a building material pose significant radiation hazard to individuals.

Key words: Ramsar, building materials, gamma spectrometry, hazard indices

Introduction

The natural radioactivity present in the environment is the main source of radiation exposure for humans and constitutes the background radiation level.[1] The main natural contributors to external exposure from gamma rays are ^{226}Ra, ^{232}Th, and ^{40}K. Since these radionuclides are not uniformly distributed, the knowledge of their distribution in soil and rocks play an important role in radiation protection and measurement. It is important to determine the sources and their individual contributions to the total radiation dose.[2] In a typical environment, respective contribution to radiation exposure is about 13.8% for ^{40}K, 55.5% for ^{226}Ra,

and 14% for ^{232}Th. The worldwide mean dose rate from terrestrial radionuclides is about 60 nGy^{-1} for areas with normal background.[3]

Life evolved in an environment with higher radiation levels than exist today, and background radiation levels today are lower than at any time in the history of life on Earth. Natural radioactivity and the respective external exposition of the population depend on the geological and geographical conditions of the region.[4-6] Areas with unusually high background radiation are found in Yangjiang, China; Kerala, India; Guarapari, Brazil; and Ramsar, Iran. The high background radiation in the Ramsar can be considered to be first due to the presence of considerable amount of ^{226}Ra along with its decay products brought to the Earth surface by numerous hot springs, radium is dissolved from the rocks by hot ground water, and second due to some travertine deposits having high thorium concentration.

Residents of these "hot" areas have used the local stone as a convenient building material to construct houses.[7,8] Building materials are the main source of indoor gamma radiation. All stone-based building materials contain radioactive nuclides, which contribute to radiation exposure. Knowledge of the level of natural radioactivity in building materials is then important to assess the possible

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radiological hazards to human health and to develop standards and guidelines for the use and management of these materials.[9]

Over the years, particular attention had been drawn to the measurement of activity levels of building materials; several authors have studied the activity concentrations of various building materials such as concrete, soil, sediment, sand, marble, granite, ceramic tiles.[10-19] The main objectives of this research is to quantify the inventories of natural radionuclides in building material samples used in Ramsar and to estimate the radiological impacts that may accrue to the dwellers in buildings constructed with these materials.

Materials and Methods

Thirty-five building material samples were collected from different parts of Ramsar (19 local building materials and 16 market available materials). The study area is situated in the north of Iran, between longitudes 50°21′ to 50°46′ East of the Greenwich meridian and latitudes 36°34′ to 36°58′ North of Equator [Figure 1].

The commonly used market available building materials were marble, granite, brick, Travertine, block, concrete, gypsum, and mosaic which were collected randomly from sites where housing was under construction. Residents of Ramsar have also used the local stone as a convenient building material to construct houses. In selection process of natural building materials, a fully portable handheld scintillometer (NaI) was used. The system was calibrated against a ^{137}Cs standard and daily checked for stability using a low activity [137]Cs (9 kBq) radioactive source.

Collected samples were first ground to fine powder, dried, weighed (300 gr) and then covered securely and placed in polyethylene cylindrical gas-tight containers three weeks prior to counting. At the end of this in-growth period, the samples were counted for 3-16 h.
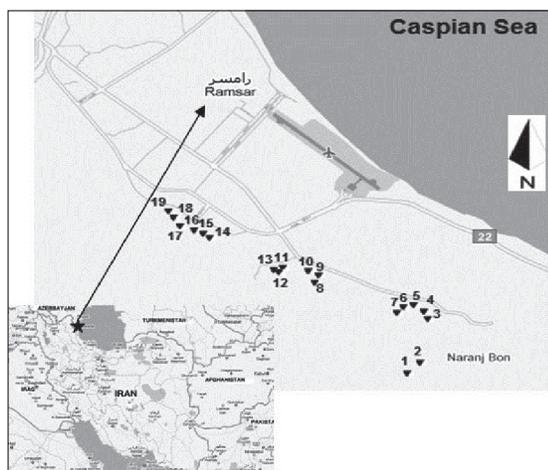

Figure 1: Study area and the distribution of local building material samples

The activity concentrations of [226]Ra, [232]Th, and [40]K in collected samples were determined using a Canberra High Purity Germanium (HPGe) detector. The detector was surrounded by a massive lead shield (10 cm) with internal walls of electrolytic copper and cadmium and coupled to a Canberra MCA-Series 100. The system was calibrated with respect to energy and efficiency using standard sources. The relative efficiency of detector is 40%, and its energy resolution is 2 keV at full width half maximum (FWHM) from 1332 keV energy of [60]Co.

Assuming secular equilibrium in the uranium and thorium decay series, the [226]Ra was determined by means of its progeny photo peak: [214]Bi (609 keV) and [232]Th was analyzed by means of its progenies photo peaks: [208]Tl (583 keV) and [228]Ac (911 keV). The activity of [40]K was measured directly through its 1461 keV peak.[20] The minimum detection limit (MDL) for [226]Ra, [232]Th, and [40]K are estimated to be 1.4, 3.04, and 2.83 Bqkg^{-1} respectively.

Results and Discussion

Activity concentrations

The measured specific activities of [226]Ra, [232]Th, and [40]K, in samples collected from different locations of Ramsar, which are organized as the frequencies of activity intervals, are presented in Figure 2. The geometric mean (GM) with its standard deviation (GSD) and the ranges of specific activities are given in Table 1.

The world average specific activity values of [226]Ra, [232]Th, and [40]K are 35, 35, and 400 Bqkg^{-1}, respectively, as reported by UNSCEAR 2000.[21] It can be observed from the results that the geometric mean specific activity of [226]Ra in the study area is much higher than the worldwide average value of 35 Bqkg^{-1}. The study can, therefore, be regarded as an area with high background radiation.

The amounts of [226]Ra, [232]Th, and [40]K for each sample are presented in our previous paper.[22]

Radiological hazard

It is important to have an assessment of the radiological hazard associated with the exposure to the radiation from [40]K, [226]Ra, and [232]Th. To account for the collective effect

Table 1: Geometric mean and range of specific activities of concerned radionuclides in building material samples collected from different areas of Ramsar

	Specific activity (Bqkg^{-1})		
	[226]Ra	[232]Th	[40]K
GM (GSD)	179.35 (14.64)	28.60 (2.63)	201.98 (4.07)
Range (min-max)	7.1–86400	4.33–187	4.44–1350

GCD: Geometric Standard Deviation

of the activity concentrations of these radionuclides in a material, a single quantity termed as "hazard index" is defined. Different hazard indices, such as Radium equivalent activity (Ra_{eq}), external radiation hazard (H_{ex}), gamma index (I_γ), and alpha index (I_γ), are used for the assessment of radiological hazard of building materials. Based on the activity concentration obtained from collected building material samples of Ramsar, and applying the appropriate formulae, hazard indices were calculated. These indices have been described in many papers.[17,23-26]
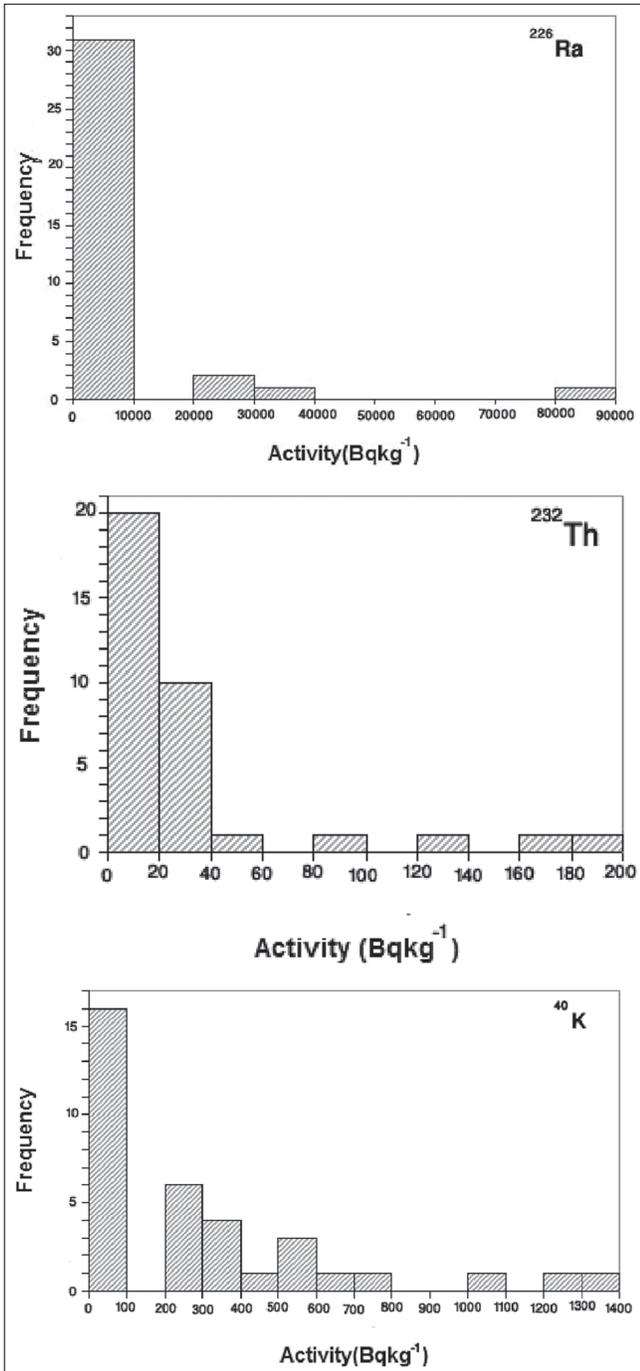

Figure 2: The frequency of measured specific activities

Radium equivalent activity (Ra_eq)

Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them. The Ra_{eq} is related to the external gamma dose and internal dose due to radon and its daughter.

The maximum value of Ra_{eq} must be less than 370 Bqkg^{-1}. This index is given as follows:[21]

$$Ra_{eq} = 370(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}) \quad (Bqkg^{-1}) \quad\quad .....(1)$$

where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The Ra_{eq} for building material samples of Ramsar ranged from 0.34 to 86400 Bqkg^{-1} with the geometric mean value of 144.80 Bqkg^{-1}. The Ra_{eq} value in some samples is more than maximum limit of 370 Bqkg^{-1} suggested by UNSCEAR for the safe use of building material. The mean and range of Ra_{eq} obtained for the analyzed samples (using equation 1) are presented in Table 2.

External Hazard Index (Hex)

The external hazard (H_{ex}) index is another radiation hazard index defined by Beretka and Mathew to evaluate the indoor radiation dose rate due to the external exposure to gamma radiation from the natural radionuclides in building materials of dwellings.[27] This index value must be less than unity to keep the radiation hazard insignificant. This index is given as follows.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \quad (Bqkg^{-1}) \quad\quad .....(2)$$

where A_{Ra}, $A_{Th,}$ and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The mean and ranges of H_{ex} for the studied samples are also given in Table 2. Results show that the values of H_{ex} in 7 of local samples are more than unity. These samples were collected from Taleshmahalleh (L1 (233.51 Bqkg^{-1}), L5 (1.13 Bqkg^{-1}), L6 (58.64 Bqkg^{-1}), Absiah L9 (26.06 Bqkg^{-1}) and Khaksefid (L7 (60.81 Bqkg^{-1}), L18 (2.27 BqKg^{-1}), L19 (98.91 Bqkg^{-1}). Values of H_{ex} in these samples indicate that the radiation hazards arising from the use of these samples is high. L1, L5, L6, L7, L9, L7, L18, and L19 are codes of local samples. See our previous paper.[22]

Table 2: Mean and range of radiation hazard indices measured in building material samples

	Radiation hazard indices			
	$Ra_{eq}(Bqkg^{-1})$	$I_\gamma(Bqkg^{-1})$	$I_\alpha(Bqkg^{-1})$	$H_{ex}(Bqkg^{-1})$
GM (GSD)	144.80 (16.3)	0.53 (15.16)	0.9 (13.27)	0.39 (16.13)
Range (min-max)	0.34–86400	0.001–288	0.03–432	0.001–233.51

GSD: Geometric Standard Deviation

Gamma Index (I_γ)

I_γ, the radiation hazard index proposed by the European Commission, can be used to estimate the gamma- radiation hazard levels typically for those associated with the natural radionuclides. This index takes into account the ways and quantities in which materials are utilized in building, and it is given as follows.[28]

$$I_r = (\frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000}) \quad (Bqkg^{-1}) \qquad .....(3)$$

where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The value of $I_\gamma \leq 0.5$ corresponds to a dose rate criterion of 0.3 mSvy^{-1}, whereas $0.5 \leq I_\gamma$ corresponds to a criterion of 1 mSvy^{-1}. Materials with $I_\gamma > 1.0$ should be avoided in building construction. They will deliver an effective dose rate higher than 1 mSvy^{-1} to the occupants of such buildings.[9,28]

The range and mean of I_γ for collected samples of building material in Ramsar are given in Table 2. The values of I_γ in 8 of local samples are more than unity. These samples were collected from Taleshmahalleh (L1 (288 Bqkg^{-1}), L5 (1.48 Bqkg^{-1}), L6 (72.33 Bqkg^{-1})), Absiah (L9 (32.16 Bqkg^{-1}), L16 (1.08 Bqkg^{-1})) and Khaksefid (L7 (75 Bqkg^{-1}), L18 (2.94 BqKg^{-1}), L19 (122 Bqkg^{-1})). Value of in these samples are more than unity and increase the annual effective dose to a member of the public by more than 1 mSvy^{-1}. And, according to the European Commission report, such materials should be avoided in building construction. L1, L5, L6, L7, L9, L16, L18, and L19 are codes of local samples. See our previous paper.[22]

Alpha index (I_α)

The excess alpha radiation due to radon inhalation originating from building materials is estimated through the alpha index (I_α), which is defined as follows.[17,27]

$$I_\alpha = \frac{A_{Ra}}{200} \quad (Bqkg^{-1}) \qquad .....(4)$$

where A_{Ra} is the activity concentration of ^{226}Ra. The recommended maximum concentration of ^{226}Ra is 200 Bqkg^{-1}, which gives $I_\alpha = 1$. When the ^{226}Ra activity concentration of any building material exceeds this maximum value (200 Bqkg^{-1}), it is possible that radon exhalation from such material could cause the indoor radon concentration to exceed 200 Bqkg^{-1}. The range and mean values of I_α obtained from the concentrations of ^{226}Ra in Ramsar building material are presented in Table 2. Values obtained for I_α in 7 of local samples are more than unity. These samples were collected from Taleshmahalleh (L1 (432 Bqkg^{-1}), L6 (108 Bqkg^{-1})), Absiah (L9 (48 Bqkg^{-1}), L16 (1.58)) and Khaksefid (L7 (112.50 Bqkg^{-1}), L18 (2.37 BqKg^{-1}), L19 (183 Bqkg^{-1})). It can, therefore, be said that radon inhalation from buildings constructed with these materials is so large as to warrant restriction of their use in building construction. There is more information about Radon exhalation rate in building materials of Ramsar in our previous study.[22]

Conclusions

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K have been measured in 35 building material samples collected from different areas in Ramsar, Iran, using gamma ray spectrometry. Results indicate that the measured mean value for ^{226}Ra specific activity is more than the world average of 35 Bqkg^{-1}. Especially 8 samples collected from Taleshmahalleh, Absiah, and Khaksefid had very high radioactivity. According to the results, Ramsar can be regarded as a region with high background radiation areas. The radiological implications of using these building materials were examined through the estimation of various radiation hazard indices and dose rates. The results in some samples were above the maximum permissible limits of these indices. It can, therefore, be concluded that an assessment of the radiological hazard of using these building materials is crucial. The use of these building materials, especially local ones with high radiation hazard indices as a result of high activity concentrations, should be restricted, and in some cases, it should be forbidden.

According to the results, the use of some local building materials in Ramsar is discouraged, and the replacement of them by building materials with low radioactivity contents is recommended.

Acknowledgment

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