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Analytical study of radionuclide concentration and radon exhalation rate in market available building materials of Ramsar

Elham Bavarnegin^{1*}, Masoud Vahabi-moghaddam¹, Asad Babakhani² and Nasrin Fathabadi²

Abstract

Samples of structural and covering market available building materials from Ramsar, a northern city of Iran, were analyzed for their radon exhalation rate using an active radon gas analyzer with an emanation container. The radon exhalation rate varied from below the minimum detection limit of 0.01 to 0.31 Bq·m⁻²·h⁻¹ with an average of 0.08 Bq·m⁻²·h⁻¹. The ²²⁶Ra, ²³²Th, and ⁴⁰K contents were also measured using a high resolution HPGe gamma-ray spectrometer system. The radionuclides contents varied from below the minimum detectable activity up to 73.5, 169, and 1,350 Bq.kg⁻¹, with the average value of 16 ± 6, 25 ± 11, and 280 ± 101 Bq.kg⁻¹, respectively. It was concluded from the results that some granite samples along with the block sample were the main source of radon exhalation rate, and the mean values of ²²⁶Ra, ²³²Th, and ⁴⁰K in building material samples are below the world average values. Therefore, the use of these market available building materials in construction of Ramsar dwellings is considered to be safe for human habitation.

Keywords: AlphaGuard 2000, Building materials, Gamma spectrometry, Ramsar, Radon exhalation, Radon-222

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Background

Natural radionuclides are present everywhere in the environment, and they constitute the greatest percentage of human exposure to ionizing radiation [1]. The main natural contributors to external exposure from gamma rays are ²²⁶Ra, ²³²Th, and ⁴⁰K. In a typical environment, their respective contribution to radiation exposure is about 13.8% for ⁴⁰K, 55.8% for ²²⁶Ra, and 14% for ²³²Th [2].

In addition to soil and water sources of indoor radon, construction materials can be a significant contributor. ²²²Rn is a daughter of ²²⁶Ra and is in turn derived from the longer-lived antecedent ²³⁸U. Since most materials contain ²³⁸U, therefore, any material can be a potential radon emitter. However, some materials have higher concentrations of ²³⁸U and ²²⁶Ra such as alum shale and black shale. Certain granites are typical of uranium-bearing natural materials, but it is always possible to find uranium-rich bedrocks of different types used locally as building

materials. Construction materials are sources of airborne radioactivity and external radiation from the decay series of uranium in buildings. Exhalation of radon (²²²Rn) from these materials is of interest since the short-lived decay products of radon are the greatest contributors to the lung dose of inhaled radionuclides [3]. Even though radon inlet into houses is a complex process involving building materials, soil, gas, water, and weather-related factors, ²²⁶Ra in construction materials may be in some cases the predominant source. Much of the radon is released from the radium trapped in the mineral grains in building materials. In building materials with high radium levels, the radon exhalation may become of major importance [4]. Areas with unusually high background (high background radiation areas (HBRAs)) are found in Yangjiang, China; Kerala, India; Guarapari, Brazil, and Ramsar, Iran. The high background radiation in the Ramsar can be considered to be due to the presence of considerable amount of ²²⁶Ra along with its decay products brought to the earth surface by numerous hot springs [5,6].

Over the years, radon exhalation from building materials has been the subject of many studies [7-12]. The main

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objective of this research is to quantify the radon exhalation rate from numerous constructions and covering building material samples used in Ramsar.

Methods

Sixteen commonly used structural and covering market available building materials were collected randomly from sites where housing and other buildings were under construction and from building material suppliers in Ramsar. 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 building materials were marble, granite, brick, travertine, block, concrete, gypsum, and mosaic. Samples were prepared in a way that their volume and surface were calculable. The surface of samples varied from 116 to 1,040 cm², and their volume varied from 120 to 82,720 cm³.

Measurement of radon exhalation rate

A radon gas analyzer-type AlphaGuard 2000 from Genitron Instruments (Germany) was used to measure radon emanation from the samples. AlphaGuard is an ionization chamber. The radon analyzer and the building material samples (Figure 2) were placed one at a time in an emanation and calibration cylindrical container

consisting of a firm corrosion-resisting stainless-steel container with a removable gas-tight lid (Figure 3). Its volume was $50.1 \times 10^{-3} \text{ m}^3$. The lid was equipped with three gas-tight electric ducts. One duct server, together with a special charger, was used as a power supply for the radon analyzer. The second duct was used to connect the fan on the middle of the inner side of the lid to the power supply by means of a power adapter. The fan was used to ensure an even distribution of the radon emanation from the sample in the interior of the container. The third duct provided communication between the gas analyzer in the interior of the container and an external PC. The concentration of radon emanated from each building material sample inside the emanation container was allowed to build up with time, and it was measured in 1 h cycles for an average time of 48 h.

The buildup of radon activity inside the emanation container follows the equation below:

$$A_t = A_0(1 - e^{-\lambda t}) \quad (1)$$

where λ is the decay constant of the nuclide concerned, and A_0 is the total value of the activity at t approximately $7 T_{1/2}$, which is approximately 27 days for radon. Via computer software program for curve-fitting the radon buildup, a sufficiently exact extrapolation of

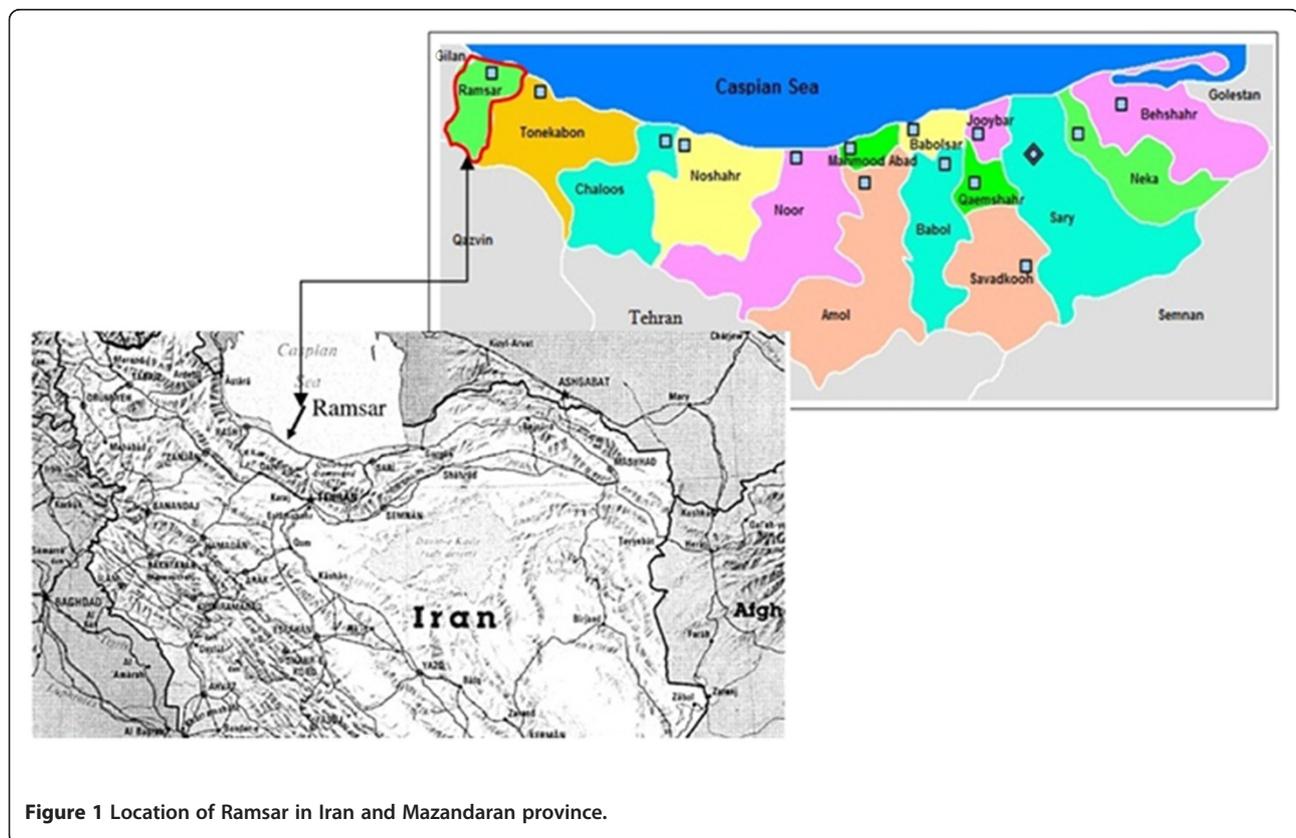


Figure 1 Location of Ramsar in Iran and Mazandaran province.

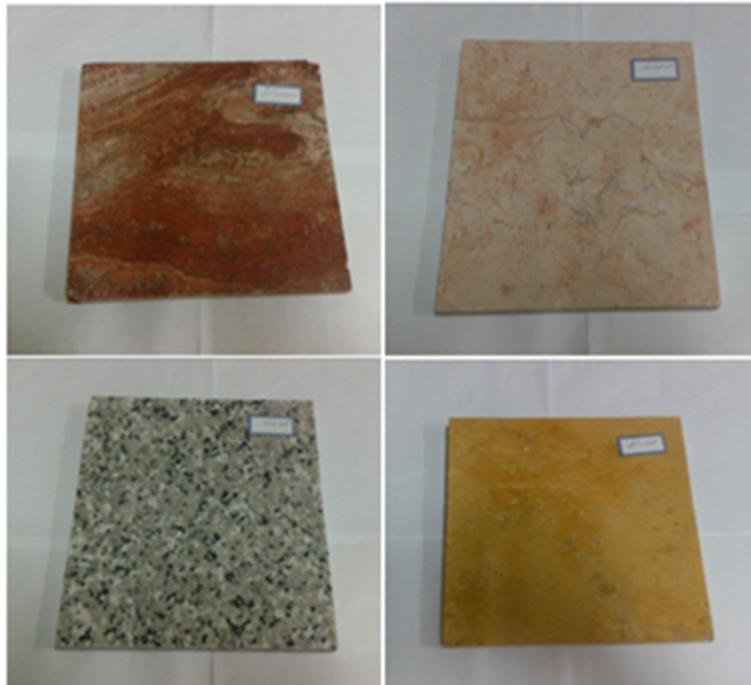


Figure 2 Some prepared samples for putting inside of calibration container to measure radon exhalation rate.

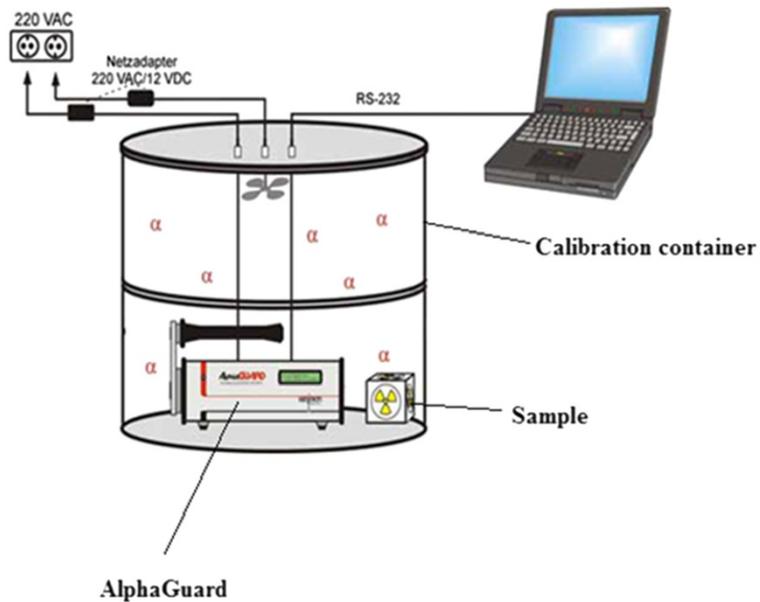


Figure 3 Schematic diagram showing the active setup used for the exhalation measurements of samples with AlphaGuard.

Table 1 Measured radionuclide concentration and radon exhalation rate in samples

| Code of market available samples | Samples type | ²²⁶ Ra (Bq.kg ⁻¹) | ²³² Th (Bq.kg ⁻¹) | ⁴⁰ K (Bq.kg ⁻¹) | E (Bq.m ⁻² h ⁻¹) |
|----------------------------------|--------------|--|--|--|---|
| M1 | Marble | < ^a MDA | < ^a MDA | < ^a MDA | ^b ND |
| M2 | Granite | 13.8 ± 0.8 | 10.2 ± 1.1 | 204 ± 6.1 | 0.07 ± 0.01 |
| M3 | Brick | 41 ± 3.4 | 31.2 ± 2.9 | 491 ± 46.2 | 0.19 ± 0.02 |
| M4 | Gypsum | 12 ± 2 | 11 ± 1.9 | < ^a MDA | ^b ND |
| M5 | Marble | < ^a MDA | < ^a MDA | 22.2 ± 12 | ^b ND |
| M6 | Marble | < ^a MDA | < ^a MDA | < ^a MDA | ^b ND |
| M7 | Travertine | < ^a MDA | < ^a MDA | < ^a MDA | ^b ND |
| M8 | Granite | < ^a MDA | < ^a MDA | 61.2 ± 13.1 | ^b ND |
| M9 | Marble | < ^a MDA | < ^a MDA | 15.1 ± 3.9 | ^b ND |
| M10 | Granite | 45.2 ± 3.7 | 83.9 ± 6.6 | 1,020 ± 91.6 | 0.28 ± 0.30 |
| M11 | Travertine | < ^a MDA | < ^a MDA | 4.44 ± 3.9 | ^b ND |
| M12 | Mosaic | 7.1 ± 0.7 | 4.33 ± 0.9 | 74.9 ± 4.6 | 0.04 ± 0.01 |
| M13 | Block | 27.0 ± 2.5 | 30.4 ± 2.8 | 297 ± 29.3 | 0.31 ± 0.03 |
| M14 | Brick | 35.7 ± 3 | 37.1 ± 3.3 | 571 ± 52.4 | ^b ND |
| M15 | Concrete | < ^a MDA | 22.5 ± 1.2 | 381 ± 8 | 0.23 ± 0.03 |
| M16 | Granite | 73.5 ± 5.6 | 169 ± 12.7 | 1,350 ± 121 | 0.24 ± 0.03 |

^aMDA, minimum detectable activity; ^bND, not detectable.

the final activity A_0 in Bq.m⁻³ was achieved, and the percentage divergence was calculated. The radon exhalation rate of samples, E , is defined as the flux of radon released at the surface of the material. This was calculated using the following formula [13]:

$$E = A_0 \lambda \frac{V}{S} \quad (2)$$

Where λ is the decay constant of radon (7.567×10^{-3} h⁻¹), V is the effective volume of calibration container (m³), and S is the surface of samples (m²).

Effective Volume = (Volume of calibration container)– (Volume of AlphaGuard + Volume of sample).

Measurement of radionuclide concentration in building materials

To determine the radionuclides concentration, samples were ground to fine powder with a particle size of less than 1 mm. Three hundred grams of samples were sealed in to a polyethylene cup for gamma spectrometry. The measurement were carried out after 4 weeks to be sure that secular equilibrium between ²²⁶Ra and its daughters had been reached. The specific activities were

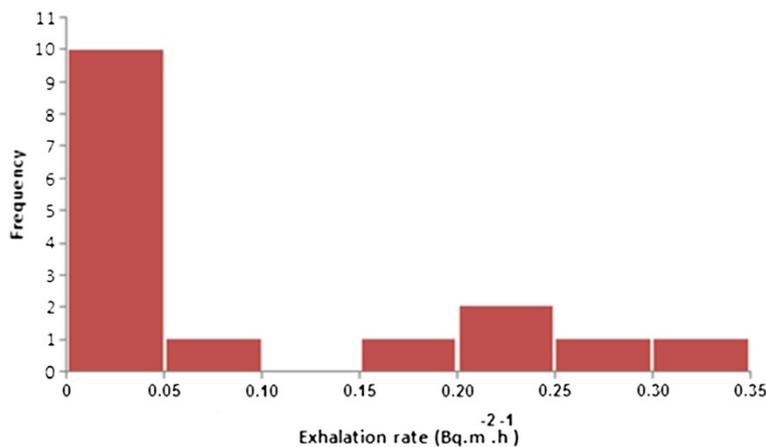


Figure 4 Frequency distribution of radon exhalation rates from samples.

evaluated by gamma-ray spectrometry on samples using a high purity germanium (HPGe) detector, that its relative efficiency was 40% with a resolution of 1.87 keV at 1.332 MeV. 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 1,461 keV peak. The minimum detectable activity (MDA) for ^{226}Ra , ^{232}Th , and ^{40}K are estimated to be 1.39, 3.04, and 28.3 $\text{Bq}\cdot\text{kg}^{-1}$, respectively. Calibration sources used were IAEA reference materials RGU-1, RGTh-1, and RGK-1.

Results and discussion

Radon exhalation rate was measured for numerous market available building materials of Ramsar using the radon gas analyzer. The radionuclide contents were also measured using a high resolution HPGe gamma-ray spectrometer system. The world average specific activity values of ^{226}Ra , ^{232}Th , and ^{40}K are 35, 30, and 400 $\text{Bq}\cdot\text{kg}^{-1}$ respectively, as reported by UNSCEAR 2000 [14]. The specific activities of ^{226}Ra , ^{232}Th , and ^{40}K in study samples varied from below the minimum detectable activity up to 73.5, 169, and to 1,350 $\text{Bq}\cdot\text{kg}^{-1}$, with the average value of 16 ± 6 , 25 ± 11 , and 280 ± 101 $\text{Bq}\cdot\text{kg}^{-1}$, respectively. The natural radionuclide contents and radon exhalation rate in samples are presented in Table 1. The radon exhalation rates varied from below minimum detection limit of 0.01 to 0.31 $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ with an average value of 0.08 $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$. Radon exhalation rate in all samples are below 0.35 $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$. Samples like travertine, marbles, gypsum, and even some granite samples did not show significant radon exhalation which indicates that they did not contain detectable traces of radium. The low exhalation of marbles supports the findings of Al-Jarallah et al. [9]. Radon exhalation rate in block is higher than other samples. It can be because of the porosity in block. The high exhalation rate in some granite samples also supports the findings of Al-Jarallah et al. [9].

Frequency distribution of radon exhalation rates from samples is also exhibited in Figure 4. According to this figure, radon exhalation rate in 62.5% of samples are below 0.05 $\text{Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$.

Conclusions

Geological materials usually contaminated with naturally occurring radioactive materials (NORM) have become a focus of great attention. These NORM under certain conditions can reach hazardous contamination levels. Some contamination levels may be sufficiently severe that precautions must be taken. Sixteen commonly market available building materials, which nowadays are

used in Ramsar, were analyzed for their natural radioactivity and radon exhalation rate. Natural radionuclides concentration in building material samples vary according to the type and origin of the building materials. Most building materials contain small amounts of NORM. The obtained results indicate that the mean of specific activities in collected samples are less than the world average of 35, 30, and 400 $\text{Bq}\cdot\text{kg}^{-1}$ for ^{226}Ra , ^{232}Th , and ^{40}K , respectively, as reported by UNSCEAR [14]. Radon exhalation rates in concrete, in some granite, and in block are more than other samples. However, marble and travertine samples indicate low radon exhalation rate.

It can be concluded as well from the results obtained in the current work that the mean values of ^{226}Ra , ^{232}Th , and ^{40}K of building material samples are all well below the world average. Therefore, the use of these building materials in construction of Ramsar dwellings is considered to be safe for human habitation. In view of the above results, these building materials are quite safe to be used for residential construction in the Ramsar.

Since only few samples of market available building materials have been investigated in the current work, local building materials, accompanying these samples, from different areas of Ramsar are also being investigated in a further study in order to get more representative values for the level of naturally occurring radioactive materials in construction materials, which will be useful for establishing a national standard on radioactivity of building materials in Ramsar [15].

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