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## **Research article**

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# Modelling of indoor external and internal exposure due to different building materials containing NORMs in the vicinity of a HNBRA in Mahallat, Iran



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Mohammademad Adelikhah<sup>a</sup>, Morteza Imani<sup>b</sup>, Miklós Hegedűs<sup>a</sup>, Tibor Kovács<sup>a,\*</sup>

<sup>a</sup> Institute of Radiochemistry and Radioecology, Research Centre for Biochemical, Environmental and Chemical Engineering, University of Pannonia, 8200 Veszprém, Hungary

<sup>b</sup> Engineering Department, Shahid Beheshti University, Tehran, Iran

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## ABSTRACT

In this study, by considering the Naturally Occurring Radioactive Materials (NORMs) contained in the building materials used in Mahallat, Iran - an area exposed to a high level of natural background radiation - residential scenarios were simulated by applying the computer code RESRAD-BUILD to estimate the long-term Effective Dose rate of three different cases of basic building materials utilized in walls, floors and ceilings. Maximum effective dose rates of between 504 and 1433  $\mu$ Sv yr<sup>-1</sup> were calculated in the second case study, tiled cement floor. The highest external and radon doses were also calculated to be 369 and 1064  $\mu$ Sv, respectively. The simulation results revealed that <sup>232</sup>Th and <sup>40</sup>K contribute the most and least to the indoor dose, respectively. As a result of a sensitivity analysis, it was found that the air exchange rate is a key variable to easily reduce the radiological impacts of building materials. It was also shown that due to the presence of <sup>226</sup>Ra, the sensitivity of effective dose to changes in wall thickness was higher than other radionuclides found in the building materials.

## 1. Introduction

Humans are continually exposed to ionizing radiation from natural radionuclides, including from terrestrial media, building materials, water, air and food. Among them, building materials contain different amounts of naturally occurring radioisotopes, mainly radionuclides from the <sup>238</sup>U and <sup>232</sup>Th decay series as well as the <sup>40</sup>K isotope, especially if they are produced using Naturally Occurring Radioactive Materials (NORMs). Hence, building materials and NORMs have more frequently been subjected to monitoring and regulation (ICRP, 1994; European Commission, 1999; UNSCEAR, 2000, 2008; Council of the European Union, 2014; IAEA, 2019). In terms of radiation and health physics, exposure to such sources of radiation is a major public health concern, especially in High Natural Background Radiation Areas (HNBRAs) such as Yangjiang in China, Guarapari in Brazil, Kerala and Madras in India, as well as Ramsar and Mahallat in Iran (UNSCEAR, 2000) where people are exposed to much greater doses of radiation than the worldwide average background dose (2.4 mSv) (UNSCEAR, 2000). In some part of Ramsar, a famous area in radiation point of view, the indoor and outdoor gamma radiation absorbed dose rates range from 50 to 900  $\mu$ Gy h<sup>-1</sup>, although other areas in the city have absorbed dose rates that are much lower. The annual dose to monitored individuals ranges up to 132 mGy, and the calculated maximum valid annual radiation exposures of up to 260 mGy (Ghiassi-Nejad et al., 2002). In Mahallat, based on the UNSCEAR, 2000, the absorbed dose rate in this area was estimated to fall within the range of 800–4000 nGy  $h^{-1}$  (including cosmic and terrestrial radiation), which is at least fifteen times higher than the worldwide average of 55 nGy  $h^{-1}$  (UNSCEAR, 2000).

Over recent years, many studies have been carried out to assess the natural radioisotopes contained in building materials in Iran (Fathivand et al., 2007; Fathivand and Amidi, 2007; Fathabadi et al., 2011; Mehdizadeh et al., 2011; Asgharizadeh et al., 2012; Bavarnegin et al., 2013; Shahrokhi et al., 2020; Imani et al., 2021) and also measure the indoor gamma dose rate all around the world (Inoue et al., 2020; Kuzmanović et al., 2020; Top et al., 2020; Tchorz-Trzeciakiewicz and Rysiukiewicz, 2021; Jeelani et al., 2021). Furthermore, two recognized HNBRAs in Iran (Ramsar and Mahallat) have also been investigated to determine the sources of radiation that affect the health of inhabitants, epidemiological risks, as well as the distribution of natural radioisotopes in different building materials (Sohrabi et al., 1996; Ghiassi-Nejad et al., 2002; Mortazavi et al., 2005; Bavarnegin et al., 2013; Shahrokhi et al., 2020; Adelikhah et al., 2020, 2021; Shahrokhi et al., 2021a,b). The primary origin of these NORMs is mainly due to the presence of a very high level of <sup>226</sup>Ra in the soil and igneous rocks in bedrock which are rich in

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<sup>\*</sup> Corresponding author. *E-mail address:* kt@almos.uni-pannon.hu (T. Kovács).

uranium in the Iranian cities of Ramsar and Mahallat (Ghiassi-Nejad et al., 2002; Shahrokhi et al., 2020). In addition, some radiological assessment models such as RESRAD-BUILD, FLUENT, RAETRAP and COMIS were applied to identify the risks found in different scenarios, and different methods were proposed for the purpose of significantly reducing the radiological hazards from building materials (Risica et al., 2001; Janssen, 2003; Abdallah et al., 2012; Akbari et al., 2013; Pepin, 2018; Abdullahi et al., 2020; Kocsis et al., 2021). On the other hand, the available information is still limited, some areas are still lacking adequate information.

Accordingly, a survey has been designed firstly, to measure the radioactivity content of the most common building materials available on the local market of Mahallat (an area exposed to a high level of natural background radiation); and secondly, to estimate the contribution of building materials towards indoor external and internal doses received by inhabitants in Mahallat using the computer code RESRAD-BUILD in a standard model room developed by Argonne National Laboratory (USA). The exposure pathways to estimate effective dose were including inhalation, radon gas, external exposure, deposition and immersion. In addition, three different case studies using different compositions of basic building materials, taking into account the different densities and thicknesses of such materials, were applied in a simulation to determine how much of an influence each one has on the effective dose rate occupants are exposed to. The impact of indoor environmental parameters on the external and internal doses was also investigated.

## 2. Materials and methods

#### 2.1. Building material sampling and preparation

The building materials studied - namely brick, cement, sand, tiles and ceramic - are commonly used by local people for construction and were randomly collected from local suppliers and/or manufacturers. Prior to conducting the radiation counting of the samples, a soil reference material sourced from the International Atomic Energy Agency (IAEA-375) with a known activity concentration of radionuclides was used to calibrate the system. The samples were transferred to the laboratory, then stored and dried at room temperature for several days before being pulverized, homogenized and sieved (grain size <3 mm) to achieve identical parameters to the reference material (IAEA-375, soil standard). Afterwards, the samples were dried in a ventilated oven at 90 °C for 24 h to reach a constant weight. 500 grams of each prepared and homogenized sample was transferred into a leak-proof Marinelli beaker, weighed and sealed for 30 days in order to reach a secular equilibrium between <sup>226</sup>Ra, <sup>222</sup>Rn and their gamma-emitting progeny (Shahrokhi et al., 2020; 2021a,b).

The concentrations of three naturally occurring radionuclides ( $^{40}$ K,  $^{226}$ Ra and  $^{232}$ Th) in the samples of building materials were determined using a High Purity Germanium (HPGe) gamma-ray detector (ORTEC GMX40-76) with a relative efficiency of 40% and an energy resolution of 1.95 keV at 1332.5 keV. The spectra were recorded by an ORTEC DSPEC LF 8196 MCA (multichannel analyzer) and analyzed using the Aptec MCA software.

#### 2.2. Exposure scenario and input parameters

In this study, the parameters of a standard model room, with the dimensions of  $4.0 \times 5.0 \times 2.8$  m outlined in the document entitled Radiation Protection 112, were applied in the simulation (European Commission, 1999). Since all buildings are constructed from a combination of basic building materials, variations in the proportions of such materials used to compose walls, floors and ceilings were applied in the simulation by taking into consideration their different densities and thicknesses to evaluate the influence of such parameters on the variation in the external and radon doses an individual in the simulated room is exposed to using the computer code RESRAD-BUILD. Ceilings and walls were built from the same basic materials - namely brick, cement and sand - in equal proportions. Three different forms of floor – that is, cement, tiled and ceramic - were also assumed. The location of the exposed person for the purpose of the external dose assessment was chosen to be at the center of the room and 1 m above the floor (Yu et al., 2000), moreover, an indoor occupancy rate of 80% was assumed (UNSCEAR, 2000). The density of the building materials was also assumed to be 2000 kg m<sup>-3</sup> for brick, 2960 for gypsum, 2400 for tiles and 2350 for cement. The simulations were performed over 76 years, the life expectancy of Iranians. Other main input parameters used as input data regarding dose assessments are also listed in Table 1:

Parameters	Unit	Value
Breathing rate	$m^3 d^{-1}$	22
Deposition velocity	$m s^{-1}$	$1 imes 10^{-2}$
Ventilation rate	$h^{-1}$	0.1–2
Radon emanation rate	-	0.2
Porosity	-	0.1
Radon diffusion rate	$m^2 s^{-1}$	$2  imes 10^{-5}$
Resuspension rate	-	$5  imes 10^{-7} \ s^{-1}$
Release air fraction	-	0.1
Type of the source	-	Volume (x-, y and z-direction)
Source geometry	-	Rectangular
Shielding thickness	cm	No shielding

Table 1. The main parameters applied for effective dose calculations.

#### 2.3. The computational code RESRAD-build 3.5

RESRAD-BUILD is a model designed to estimate the possible radiation dose received by an individual, who works or lives in a building containing radioactive material, by taking into account different pathways for human exposure including external exposure directly from the source, external exposure due to air submersion and internal exposure due to indoor <sup>222</sup>Rn, <sup>220</sup>Rn, their progenies, etc. This code allows users to calculate the time-integrated dose over the period of exposure at time intervals specified by the user (Yu et al., 2003). The simulation of each exposure pathway was carried out using several equations, e.g., the total external dose ( $D_{ext}$ ) at time *t* that occupants are exposed to over the exposure duration is expressed as (Yu et al., 2003):

$$D_{ext}(mSv y^{-1}) = \frac{ED}{365} \times F_{in} \times F_i \times C \times DCF \times F_g$$
(1)

where *ED* denotes the exposure duration in days (d) and  $F_{in}$  as well as  $F_i$  represent fractions of time spent indoors and in the dimensionless room *i*, respectively. *C* stands for the mean activity concentration of the radionuclide (Bq g<sup>-1</sup>); *DCF* refers to the dose conversion factor (mSv y<sup>-1</sup> (Bq g<sup>-1</sup>)<sup>-1</sup>) which is one of the in-built dose libraries in the RESRAD-BUILD computer code (ICRP 72-adult); and  $F_g$  is the geometrical factor for the source parameters including shielding, thickness, position of the occupant relative to the radionuclide source and finite area (Yu et al., 2003) that are some of the inputs to RESRAD-BUILD in light of the source parameter.

In the case of the inhalation dose of radon, an estimation of the RESRAD-BUILD computer code is made from the air quality model to evaluate the concentrations of the radon decay products by considering the exchange of air between inside and outside of the compartments as well as radioactive decay and ingrowth. The concentrations of the radon isotopes are calculated by Eq. (2), while estimation of the airborne short-lived decay products indoors is based on a model found in the literature (Jacobi, 1972) that has been extended (Porstendörfer, 1994):

$$\frac{dC_{Rn}}{dt} = \frac{EA}{V} - (\lambda + \vartheta)C_{Rn} + \vartheta C_{Rn0}$$
<sup>(2)</sup>

where  $C_{Rn}$  represents the indoor radon concentration (Bq m<sup>-3</sup>), *E* denotes the radon exhalation rate (Bq m<sup>-2</sup> h<sup>-1</sup>), *A* stands for the room area (m<sup>2</sup>), *V* refers to the volume of the room (m<sup>3</sup>),  $\lambda$  is the decay constant of radon (0.007 h<sup>-1</sup> for <sup>222</sup>Rn),  $\vartheta$  denotes the air exchange rate (h<sup>-1</sup>) and  $C_{Rno}$ stands for the outdoor radon concentration (Bq m<sup>-3</sup>).

### 3. Results and discussion

#### 3.1. Natural radioactivity in building materials

The mean activity concentrations of naturally occurring radionuclides for cases 1, 2 and 3 used in the simulation are given in Table 2 which was reported in detail in our previous work (Shahrokhi et al., 2020).

#### 3.2. Radiological dose assessment using RESRAD-BUILD computer code

Since most residents spend 80% of their lifetime indoors, the natural radioactivity emitted from walls, floors and ceilings could have a significant effect on them. The simulation and calculation of doses that humans are exposed to in a contaminated building, alternatively, because of contaminated furniture or equipment found in the building, were carried out by the computer code RESRAD BUILD. The RESRAD-BUILD computer code was developed on the basis of indoor building surface contamination and modelling of the release mechanisms, such as diffusion (radon gas and tritiated water), mechanical removal (decontamination activities), or erosion (removable surface contamination), and transport of contaminants in the indoor environment which is calculated with an indoor air quality model. The air quality model evaluates the transport of radioactive dust particulates and radon progeny due to (1) air exchange between compartments and with outdoor air, (2) the deposition and resuspension of particulates, and (3) radioactive decay and ingrowth.

As previously mentioned, several scenarios were assumed and applied in the simulation in order to determine which parameters and possible combinations of building materials have more of an influence on the effective dose rate inhabitants in Mahallat - an area exposed to a high level of natural background radiation - are exposed to. In the first round of simulations the variation in the composition of building materials making up walls, floor and ceiling was considered. Then the effect of variation in the room parameters, namely wall thickness and ventilation rate were assessed. The simulation of indoor doses including indoor external and internal exposure doses, as well as the Effective Dose was carried out over 76 years, the average lifetime of Iranians. Therefore, the measured activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were individually inputted into the RESRAD-BUILD computer code. **Figure 1** illustrates the standard room model that was simulated by the code.

Variations in effective dose from both the walls and floor of the room that occupants are exposed to are shown in Figure 2 (A-C) for a standard room with a wall thickness of 20 cm and air exchange rate (ACH) of 0.5  $h^{-1}$ . As reported by Yoshino et al. (2004), it should be mentioned that the

<b>Table 2.</b> Activity concentrations of $2^{20}$ Ra $2^{32}$ Th and $^{40}$ K for the three	ree cas	the thre	for th	nd <sup>40</sup> K	<sup>32</sup> Th	of <sup>226</sup> Ra	concentrations	Activity	2.	Table
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Activity	Case I		Case II	Case II		Case III	
	Walls and Ceiling	Cemented Floor	Walls and Ceiling	Cement floor tiled	Walls and Ceiling	Cement floor-with ceramic	
<sup>226</sup> Ra	32.0	34.0	32.0	40.0	32.0	34.0	
<sup>232</sup> Th	26.5	25.0	26.5	29.0	26.5	26.5	
<sup>40</sup> K	253.0	229.0	253.0	284.0	253.0	272.5	

ACH was assumed to be  $0.5 \text{ h}^{-1}$  in line with the minimum ACH legislated in most European countries and in accordance with Japanese regulations for ACH (Yoshino et al., 2004). Figure 2 (A-B) shows a long-term variation in the indoor external and radon doses inhabitants are exposed to over an average lifetime of 76 years for the 3 different case studies. Accordingly, a considerable rise in the indoor doses was observed over the first 30 years, however, beyond this period, the indoor doses became relatively constant. Based on Figure 2C, the maximum effective dose rate was exhibited by the case study two, namely a tiled cement floor, that ranged from 504 to 1433  $\mu$ Sv yr<sup>-1</sup>, which is below the recommended maximum limit set by the International Commission on Radiological Protection (ICRP) of 3–10 mSv y<sup>-1</sup> (ICRP, 1994). The highest external and radon doses - calculated to be 369 and 1064 µSv, respectively - were also exhibited in case study two because of the higher concentrations of <sup>226</sup>Ra and <sup>232</sup>Th in these materials. Therefore, the choice in building materials has a noticeable contribution towards the indoor doses inhabitants are exposed to.

Figure 3 (A-D) represents the fractional contribution of each radionuclide in terms of the long-term variation in indoor doses. The simulation results revealed that <sup>232</sup>Th and <sup>40</sup>K contribute the most and least towards the indoor dose, respectively. Although the indoor doses due to exposure to <sup>226</sup>Ra and <sup>232</sup>Th showed a significant degree of variation over the average lifetime of 76 years, variations are distinguishable, for example, Figure 3A shows that the relationship exhibited concerning doses as a result of exposure to <sup>226</sup>Ra is linear and significantly reduces over the average lifetime of 76 years, while Figure 3C illustrates that indoor doses as a result of exposure to <sup>232</sup>Th rose considerably over the first 30 years but were relatively stable from then on. On the contrary, from Figure 3B no variation can be seen in the indoor dose as a result of exposure to <sup>40</sup>K and, according to the simulation results, <sup>40</sup>K does not contribute to the indoor internal dose, therefore, the external dose is the contribution of the <sup>40</sup>K to the indoor doses.

#### 3.3. Sensitivity analysis

Sensitivity analysis is a technique used to study and identify the parameters that have a significant effect on the results of dose assessments and to explore potential reductions in the uncertainties associated with such parameters. In this paper, in order to more realistically estimate radiological risks, variations in essential parameters of the room, e.g., wall thickness and air exchange rate, used in the RESRAD-BUILD computer code were taken into account. In this regard, the wall thickness and air exchange rate were varied from 10 to 60 cm and 0.05 to 2.0 h<sup>-1</sup>, respectively. These parameters were applied to the materials used in case study two, as the maximum effective dose rate was regarded as a worst-case scenario as is shown and described in Figure 2.

Alterations made to the indoor doses regarding the wall thickness and air exchange rate are illustrated in Figures 4A and B, respectively. Accordingly, it can be seen that the wall thickness and air exchange rate both play a large role in terms of the indoor dose occupants are exposed to. Increasing the thickness of any part of the room means increasing the amount of the building material and NORM concentrations which will lead to a further increase in the indoor dose rate. Based on Figure 4A, as the wall thickness increases, the indoor doses also increase, moreover, for wall thicknesses of 30 cm or more, dose remains relatively constant, which is in good agreement with results presented in previous studies (Majid et al., 2013; Risica et al., 2001) and is contradictory to earlier expectation that the increase in concrete thickness would increase the dose rates in dwelling. Indeed, the dose increases with the wall thickness to a point because of the external gamma dose rate, which is mitigated by self-absorption, and the greater wall thickness also increases the potential amount of radon isotopes exhaled into the room, mitigated by the distance radon is able to travel in the building material. Consequently, when the thickness is bigger than 30 cm, it will start to serve as a shield to its own radiation and showed in Figure 4a and any



Figure 1. Standard room model simulated by the RESRAD-BUILD computer code.



**Figure 2.** Long-term variation in A) external radiation dose, B) radon dose and C) Effective dose that inhabitants are exposed to in a standard room in which the wall thickness is 20 cm and ACH is  $0.5 h^{-1}$  in three different case studies.



**Figure 3.** Long-term variation in the effective dose inhabitants are exposed to due to A)<sup>226</sup>Ra, B)<sup>232</sup>Th, C)<sup>40</sup>K and D)<sup>210</sup>Pb in a standard room with a wall thickness of 20 cm and ACH of 0.5 h<sup>-1</sup> in three different case studies.



**Figure 4.** Effect of A) wall thickness and B) air exchange rate on the effective dose inhabitants of Mahallat, Iran are exposed to.

addition of wall thickness over 30 cm would not increase further the indoor dose rates and could be ignored.

However, a continuous increase in doses as a result of wall thicknesses in excess of 40 cm and exposure to  $^{226}$ Ra found in building materials has been reported (Abdullahi et al., 2020). On the other hand, radon, one of the biggest contributors to the total dose, can easily escape from the wall matrix if the wall is narrower. If the wall is thicker, the relative amount of radon that escapes from the building decreases.

According to Figure 4B, by increasing the air exchange rate, the effective dose is dramatically reduced with values ranging from 3590  $\mu$ Sv y<sup>-1</sup> for ACHs of 0.05 h<sup>-1</sup> to 296 for ACHs of 2.00 h<sup>-1</sup>. Consequently, in terms of the indoor environment - an extremely important subject from the perspective of human health - that is, indoor air quality and energy efficiency, the ventilation rate significantly affects the inhalation dose, which is inversely correlated with radon and the airborne concentrations of its decay products.

Figure 5 A-D also show the effect of wall thickness on the effective dose attributed to each of the measured natural radionuclides with regard to the second case study at an ACH of  $0.5 \text{ h}^{-1}$ . The dose increases with the wall thickness to a point because of the external gamma dose rate, which is mitigated by self-absorption. In Fig A-B, the greater wall thickness also increases the potential amount of radon and thoron exhaled into the room, respectively, mitigated by the distance radon and thoron (which is much smaller than radon) is able to travel in the building material. Also, in Figure 5D, due to the low gamma energy of <sup>210</sup>Pb, after a slight early increase most of the gamma dose is absorbed in the material itself. Accordingly, the effective dose varied from 230 to 945 and 5–909  $\mu$ Sv y<sup>-1</sup> due to the presence of <sup>226</sup>Ra and <sup>232</sup>Th, respectively, while the effective dose due to  ${}^{40}$ K varied between 67 and 95  $\mu$ Sv y $^{-1}$ . A wall thickness of 10 cm is associated with the minimum effective dose from natural radionuclides, moreover, the highest recorded values for the effective dose due to <sup>232</sup>Th and <sup>40</sup>K resulted from a wall thickness of 30 cm and due to <sup>226</sup>Ra from a wall thickness of 60 cm.



Figure 5. Effect of wall thickness on the effective dose due to A)<sup>226</sup>Ra, B)<sup>232</sup>Th, C)<sup>40</sup>K and D)<sup>210</sup>Pb inhabitants of Mahallat, Iran are exposed to.

### 4. Conclusion

Since building materials contain different amounts of radionuclides from the decay series of  $^{238}$ U and  $^{232}$ Th and from the isotope  $^{40}$ K - the analysis of exposure inhabitants are subjected to resulting from direct external radiation (namely from radionuclides in the walls, floor and ceiling and submersion in air containing airborne particles), the inhalation of airborne contaminated dust particles, inhalation of radon and incidental ingestion of contaminated dust particles is necessary. Therefore, in this study, an attempt has been made to estimate the contribution of building materials towards indoor external and internal doses as the main pathways into inhabitants in the vicinity of HNBRA in Mahallat, Iran. Simulations were carried out using the computer code RESRAD-BUILD and a pathway analysis model designed to evaluate the potential radiological dose incurred by an individual who lives in a building contaminated with radioactive material in Mahallat. Iran. In this regard, three case studies were assumed using different building materials. The maximum long-term Effective Dose rate was recorded from the case study two (tiled cement floor), which varied from 504 to 1433  $\mu$ Sv yr<sup>-1</sup> and was below the recommended maximum limit set by the International Commission on Radiological Protection (ICRP). The highest external and radon doses were also estimated to be 369 and 1064 µSv, respectively. According to results from the simulation, the contribution of <sup>232</sup>Th towards the indoor doses is higher than that of <sup>226</sup>Ra due to the homogenous condition of both 222Rn and 220Rn in the standard model of the room simulated by the computer code RESRAD-BUILD, whereas the homogeneity of <sup>220</sup>Rn in a room is highly uncertain due to its short halflife of 56 s. Furthermore, it was also determined that the main contributor towards the variation in radon concentration is as a result of changes to the air exchange rate. Lastly, since the RESRAD-BUILD considers the decay of radionuclides and the ingrowth of decay products, the dose might appear increase due to the ingrowth of <sup>232</sup>Th decay products, while <sup>226</sup>Ra is not causing a problem due to reaching equilibrium much faster, nor does the <sup>40</sup>K because of the long half-life. Furthermore, building material would not contain pure <sup>226</sup>Ra or <sup>232</sup>Th, although they can have a disturbed decay chain.

The results indicate that the computer code RESRAD-BUILD is a useful tool to evaluate radiological human health risk for buildings contaminated with radionuclides and to monitor and control the radioactivity of building materials. The data in this study could also contribute towards the database of natural radionuclides found in building materials, improve current technical regulations and laws concerning the radioactive content of building materials, lead to the proposal of a radiological reduction method as well as raise public awareness of radiological risk. Consequently, the legislation of a national standard into the Iranian legal system describing the requirements for the radiological examination of the building materials, is necessary before their introduction on the market.

#### Declarations

## Author contribution statement

Tibor Kovács & Miklós Hegedűs: Analyzed and interpreted the data; Wrote the paper.

Mohammademad Adelikhah: Conceived and designed the experiments.

Morteza Imani: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data.

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## Data availability statement

Data included in article/supplementary material/referenced in article.

## Declaration of interests statement

The authors declare no conflict of interest.

## Additional information

No additional information is available for this paper.

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