DETERMINATION OF RADON AND NATURAL RADIOACTIVITY CONCENTRATION IN SOME BUILDING MATERIALS USED IN IZMIR, TURKEY

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ABSTRACT

The aim of this study is to determine the radon and natural radioactivity concentrations of some building materials and to assess the radiation hazard associated with those mortar materials when they are used in the construction of dwellings. Radon measurements were realized by using LR-115 Type 2 solid state nuclear track detectors. Radon activity concentrations of these materials were found to vary between 130.00 ± 11.40 and 1604.06 ± 40.5 Bq m^{-3}. The natural radioactivity in selected mortar materials was analyzed by using scintillation gamma spectroscopy. The activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K for the studied mortar materials ranged from ND to 48.5 ± 7.0 Bq kg^{-1}, ND to 41.0 ± 6.4 Bq kg^{-1} and ND to 720.4 ± 26.8 Bq kg^{-1}, respectively. Radium equivalent activities, external and internal hazard indexes, gamma and alpha indexes and absorbed gamma dose rates were calculated to assess the radiation hazard of the natural radioactivity in studied samples. The calculated Ra_{eq} values of all samples were found to be lower than the limit of 370 Bq kg^{-1} set for building materials. The estimated hazard index values were found to be under the unity and the absorbed dose rate values were also below the worldwide average of 84 nGy h^{-1}.

KEYWORDS

Radium, activity, solid state nuclear track detector, hazard index, absorbed dose rate

1. INTRODUCTION

Concrete or mortar is a building material which is commonly used everywhere. Concrete is a mixture of cement, water, aggregate (coarse and fine) and admixture (Neville, 1996). The use of Portland Cement (PC) in the concrete production may no longer be considered as sustainable due to PC containing high energy and raw materials consumed during its production. The use of environmentally friendly materials is one of the possible ways to encourage a sustainable

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approach and to lower carbon footprint. That can be realized with the use of recycled materials such as recycled aggregate and PC replacement through supplementary cementing materials such as ground granulated blast furnace slag (GGBFS), fly ash (FA), silica fume (SF) (Wang et al., 2017; Bostanci et al., 2016). Nevertheless, building materials used in the construction of dwellings may contain radioactive components in various proportions depending on their provenance. Soil and building materials are the main sources of indoor radon (Nazaroff and Nero, 1988; Righi and Bruzzi, 2006; Kovler, 2017). Following the soil, building materials are considered as the second major source of indoor radon (Kumar et al., 2014).

Radon is a colorless, odorless, tasteless and invisible radioactive gas formed naturally from the decay of radioactive elements, such as radium, which is found in different amounts in soil and rock around the world. As a member of the radioactive decay series, radon gas moves from the earth crust to the atmosphere by diffusion and advection mechanisms. The diffusion is defined as the random movement of molecules while a liquid or gas accompany radon in the advection process. Radon concentrations in buildings are dependent on the local geology and might vary notably among different parts of a country and even from building to building in the same area (IAEA, 2003). Radon is an alpha-emitting radioactive gas. It is a daughter product of ^{226}Ra and decays with a half-life of 3.82 days emitting alpha particles with energy as 5.49 MeV. As radon itself decays, it produces new radioactive elements called radon daughters or decay products. Unlike radon which is in gas formation, these decay products are solids and stick to surfaces in ambient air. If radon and its daughters are inhaled or breathed, daughter products of radon can stick to the inner walls and membranes of the human respiratory system and increase the risk of lung cancer development (Khan et al., 2012).

It was reported that members of the U and Th radioactive decay series together with ^{40}K are responsible for the main contributions to the dose from natural radiation. Building materials contain various amounts of these radionuclides. Hence, it is important to measure the activity concentration of radionuclides in building materials during the assessment of population exposures as those materials are currently used in construction and most individuals spend 80% of their time indoors (Merle and Enn, 2012; Shoeib and Thabayneh, 2014; Kayakökü et al., 2016).

Naturally formed radionuclides in building materials are the sources of external radiation exposure in dwellings. This radiation results from the gamma radiation originating from the uranium and thorium series and from ^{40}K. It is important to study the radiological characteristics of these building materials for assessing the radiation hazards arising from their use in the construction of dwellings accurately. It is also important to set standards and guidelines for the use of these materials. The worldwide average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were determined as 50, 50 and 500 Bq/kg for building materials, respectively (UNSCEAR, 1993). The population-weighted average of indoor absorbed dose rate in air from the terrestrial source of radioactivity is estimated to be 84 nGy h^{-1} (UNSCEAR, 2000) and the worldwide average indoor effective dose due to gamma rays from building materials is estimated to be 1 mSv y^{-1} (EC, 1999). The detail of all unit abbreviations is given in Appendix A, Table A1.

In this paper, radon content, natural radioactivity and the radiological effect caused by this radioactivity of commonly used mortar materials were investigated.

2. MATERIALS AND METHODS

Commonly used mortar materials for producing mortar were analyzed in terms of radon activity content and natural radioactivity. Because radon variably changes with the local geology of the studied region, 5 samples of each material were collected from different sites and their radioactivities were analyzed in a preliminary study and samples showing more potential reactivity were chosen in this study to evaluate the radiological effect caused by the selected materials. The samples were dried at 105°C during 24 hours, sieved through a 100-mesh sieve.

"Sealed Can Technique" was used for radon activity concentration measurements. For the measurements, equal amounts from each sample (200 g) were placed in the cylindrical glass jar containers. In each glass jar, a piece of LR-115 Type II solid state nuclear track detector (2 cm x 2 cm) was fixed at the top inside the cylindrical glass jar. The sensitive lower surface of the detector was freely exposed to the emergent radon in order to be able to record the tracks of alpha particles resulting from the decay of radon in the remaining volume of the glass jar. Radon and its daughters reach an equilibrium concentration after a week or more and hence the equilibrium activity of emergent radon could be obtained from the geometry of the can and time of exposure. After exposure for 4 weeks, the detectors were etched in 2.5 N NaOH solutions at 60°C for a period of 90 min in a constant temperature water bath to reveal the tracks registered in the films (Mahur et al., 2008). After etching, the detectors were washed in distilled water and dried in air. Alpha track density was determined by using a LEICA DM 750 optical microscope with 100x magnification, the digital camera LEICA ICC50 (3 Mega pixels CMOS sensor) and LEICA Application Suite (LAS EZ) coupled to a PC. Counted track density was converted to radon activity concentration by using the calibration factor of 0.0217 kBq m^{-3} (track cm^{-2} $d^{-1})^{-1}$ obtained from an earlier calibration experiment (Yarar et al., 2014). Radon concentration and radiometric measurements were performed at the Institute of Nuclear Sciences of Ege University, İzmir, Turkey.

For radiometric analysis, samples were dried for 24h in the oven at 105°C to remove moisture. Each sample was packed in 100 cm^3 polyethene cylindrical containers and sealed for 40 days to attain a long-term equilibrium between ^{238}U, ^{232}Th and their daughter products before gamma spectrometric analysis. The radionuclide activity concentrations were measured in building material samples by using a low-level gamma-ray spectrometric set-up at the Nuclear Sciences Institute of Ege University. The detector consists of a 3x3 inch NaI (T1) gamma scintillation detector coupled to a Canberra AMP/TSCA (Model 2015A) Amplifier, Canberra Multiport II and Genie 2000 spectroscopy software. The sealed sample was placed in the protection unit of the gamma-ray spectrometer for 3h (10800 s) of counting time. The ^{226}Ra activity measurement was based on 1.76 MeV gamma rays from ^{214}Bi. The ^{232}Th activity determination was based on 2.62 MeV gamma rays from ^{208}TI. The activity of ^{40}K was determined through its 1.46 MeV gamma rays (Iqbal et al., 2000; Chiozzi et al., 2000; Bolca et al., 2007; Narayana et al., 2007; Tabar et al., 2013; Saç et al., 2014; İçhedef et al., 2015). Calculation was performed three times for each sample and the arithmetic mean was determined. The system calibration was carried out with standard samples for ^{226}Ra (118ppm), ^{232}Th (600 ppm) and ^{40}K (52.45%). The detector efficiency was determined with the same standard samples. The used detector carried a photopeak efficiency of 2.6% for ^{226}Ra, 2.3% for ^{232}Th and 3.1% for ^{40}K (İçhedef et al., 2015).

3. RESULTS AND DISCUSSIONS

3.1 Radon Activity Concentration

The radon activity values of the studied samples were obtained by multiplying the track density of the etched detectors and the calibration factor equal to 0.0217 kBq m^{-3} (track cm^{-2} $d^{-1})^{-1}$ obtained from an earlier calibration experiment (Yarar et al., 2014). The values of radon activity

TABLE 1. Radon activity concentration of selected mortar materials.

Sample Type	Radon activity (Bq/m^3)
Cement (CEM I)	164.92 ± 12.84
Silica Fume	130.00 ± 11.40
Fly ash	302.06 ± 17.40
Limestone fine aggregate	204.00 ± 14.28
Basalt fine aggregate	1604.06 ± 40.5

in building material samples are shown in Table 1. It can be seen from Table 1 that the radon activity concentrations of the studied samples vary from 130.00 ± 11.40 to 1604.06 ± 40.5 Bqm^{-3}. The reference radon level given by ICRP-126 (2014) is between 100 and 300 Bq/m^3. The radon concentrations of cement (CEM I) and silica fume were within this range while it was observed that the measured radon concentration was on the upper level of reference. On the other hand, a considerably high radon level was obtained from basalt samples.

3.2 Radium, Thorium and Potassium Activity Concentrations

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are shown in Table 2. The concentration in studied mortar material samples for ^{226}Ra, ^{232}Th and ^{40}K varied from ND to 48.5 ± 7.0 Bq kg^{-1}, ND to 41.0 ± 6.4 Bq kg^{-1}and ND to 720.4 ± 26.8 Bq kg^{-1}, respectively (Table 2). The minimum detectable activity (MDA) of gamma spectrometer system for ^{226}Ra, ^{232}Th and ^{40}K are calculated according to Curie (1968) as 11 ± 3 Bq/kg, 9 ± 3 Bq/kg and 54 ± 7 Bq/kg, respectively. As seen from Table 2, all radionuclide concentrations are below the detection limits for limestone fine aggregate samples. Excluding limestone fine aggregate, ^{226}Ra concentrations of cement (CEM I), fly ash and basalt fine aggregate are found in a narrow range between 31.0 ± 5.5and 48.5 ± 7.0 Bq/kg. Additionally, ^{226}Ra concentration is not measured for silica fume due to its low concentrations. Results show that radionuclide concentrations of fly ash and basalt samples are comparable. A literature comparison related to the radionuclide activity concentrations of some building materials is given in Table 3.

TABLE 2. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in selected mortar materials.

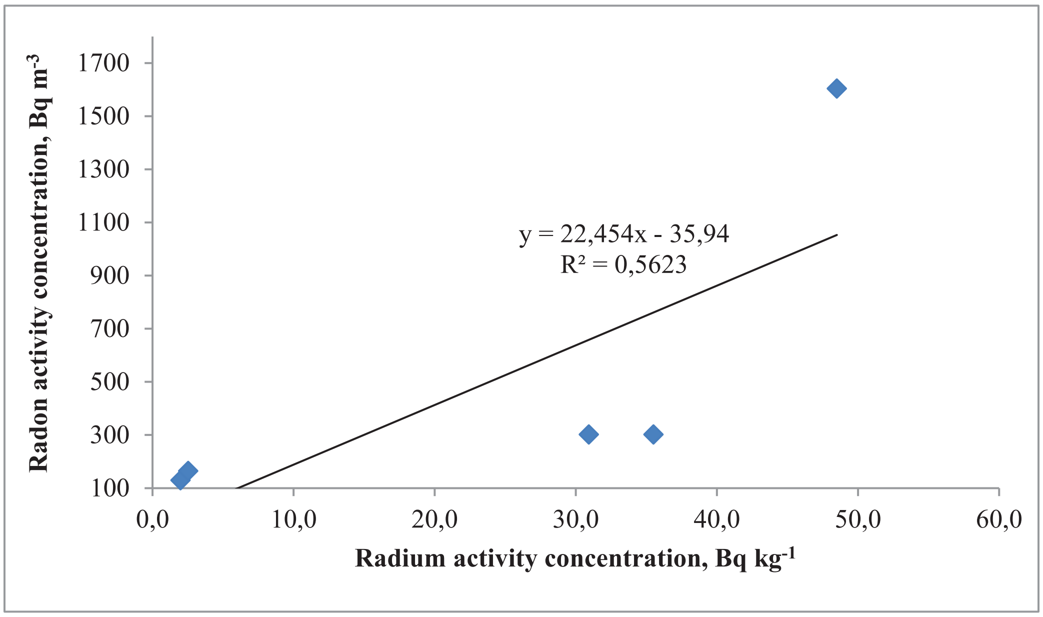
Sample type	^{226}Ra(Bq/kg)	^{232}Th(Bq/kg)	^{40}K (Bq/kg)
Cement (CEM I)	31.0 ± 5.5	14.3 ± 3.8	155.8 ± 12.5
Silica Fume	ND	31.4 ± 5.6	352.6 ± 18.7
Fly ash	35.5 ± 6.0	41.0 ± 6.4	720.4 ± 26.8
Limestone fine aggregate	ND	ND	ND
Basalt fine aggregate	48.5 ± 7.0	34.1 ± 5.8	372.0 ± 19.3

TABLE 3. Comparison of the radionuclide activity concentrations (Bq/kg) of some building materials obtained from the literature.

Country	Material	^{226}Ra	^{232}Th	^{40}K	Reference
Algeria	Red clay brick	65	51	675	Amrani andTahtat (2001)
Turkey	Brick	15.7	3.8	201.4	Baykara et al. (2011)
Turkey	Cement	24.7	20.7	2493	Baykara et al. (2011)
Turkey	Limestone	3.5	9.3	484	Baykara et al. (2011)
Turkey	Perlite	228.2	95.5	624.4	Kayakökü et al. (2016)
Pakistan	Brick	36.9–52	52.5–68	680–784	Khan et al. (2002)
Turkey	Fly Ash	187–563	65–136	305–831	Özden et al. (2017)
Turkey	Limestone	2.6–48.2	0.8–27.2	4–354	Turhan (2008)
Turkey	Fly Ash	66.6–415.0	47.6–232.0	220.9–735.5	Turhan (2008)
Turkey	CEM I	13.8–58.5	6.7–43.9	109.1–447.1	Turhan (2008)

A moderate positive correlation was found between radium activity which was determined by using a low-level gamma-ray spectrometric system and radon activity concentration determined with the sealed can technique (LR-115). The correlation between radium and radon activity concentration is given in Figure 1.

FIGURE 1. Linear regression of radium and radon activity concentration.



3.3 Radium Equivalent Activity Calculation

The distribution of natural radionuclides in studied samples is not standardized in general. In order to account for the radiation hazards associated with ^{226}Ra, ^{232}Th and ^{40}K, a common radiological index called radium equivalent activity (Ra_{eq}) was introduced by Beretka and Mathew (1985), Veiga et al. (2006) and Sharaf and Hamideen (2013). Along with assuming that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rate (UNSCEAR, 1982), Ra_{eq} is calculated by using the Eq. 1:

$$\mathrm{Ra}_{\mathrm{eq}}\left(\mathrm{Bq\ kg^{-1}}\right) = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad [1]$$

C_{Ra}, C_{Th} and C_K are the activity concentrations (Bq kg^{-1}) of radium, thorium and potassium in the samples. Construction materials with an average Ra_{eq} value less than 370 Bq kg^{-1} may be used for dwellings (OECD, 1979; Somlai et al., 1998). Radium equivalent values of the studied samples are presented in Table 4. The radium equivalent activities were found to vary from 63.35 ± 8.0 to 149.62 ± 12.23 Bq kg^{-1}. It can be observed from Table 4 that the Ra_{eq} values of the construction materials are determined to be within world standards (370 Bq/kg).

3.3 External and Internal Hazard Indexes

Beretka and Mathew (1985) reported that a radium equivalent activity of 370 Bq kg^{-1} in building material produced an external gamma radiation dose rate of 1.5 mSv/year. In order to limit the radiation dose to 1.5 mSv/year, a model based on the dose criterion was proposed by Hayabbu et al. (1995) to calculate the external hazard index (H_{ex}) as (Eq. 2):

$$\mathrm{H}_{\mathrm{ex}} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \quad [2]$$

C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bq/kg.

The value of the external hazard index is required to be less than the unity in order to keep the radiation hazard insignificant (Rati et al., 2010). Table 4 shows that the calculated external hazard index values range from 0.08 to 0.17. It can also be said that the external hazard index values for all analyzed samples are less than the unity.

In addition to the external hazard, internal exposure to radon and its short-lived decay product are also threats to the respiratory system. For that reason, a second criterion formula is suggested by Krieger (1981) to reduce the acceptable maximum concentration of ^{226}Ra to halve the normal limit (Turhan et al., 2007). The criterion is called the internal hazard index (H_{in}) and defined by the below formula (Eq. 3):

$$\mathrm{H}_{\mathrm{in}} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \quad [3]$$

For the safe use of materials under investigation in the construction of dwellings, H_{in} is also required to be less than the unity (Abbady et al., 2006). As seen in Table 4, calculated H_{in} values range from 0.19 to 0.50 and the maximum value is observed in the fly ash sample, on the other hand, the minimum value is observed in silica fume. H_{in} values of all analyzed samples are less than unity.

3.4 Radioactivity Level Index

Another radiation hazard index is the gamma radiation hazardous levels (I_γ) associated with the natural radionuclides in the mortar material samples. The radioactivity level index is required to be less than 1; otherwise, the external doses exposed to individuals exceed acceptable levels. The level of γ radiation hazard associated with the natural radionuclides in specific construction materials was estimated by EU Directive (CE, 2014) as (Eq. 4):

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad [4]$$

The radioactivity level index values are presented in Table 4 where the lowest value (0.23) was found in cement (CEM I) while the highest value (0.56) was found in the fly ash sample. The radioactivity level index of all studied samples is below the recommended value for a mortar material to be used in the construction of dwelling except for the fly ash sample.

3.5 Alpha Index

The excess alpha radiation caused by inhalation of radon liberated from the building materials can be estimated using the alpha index (I_α) which has been determined by Eq. 5 (Rafique et al., 2011).

$$I_\alpha = \frac{C_{Ra}}{200}\ \text{Bq kg}^{-1} \quad [5]$$

The recommended exemption and recommended upper levels of ^{226}Ra concentrations in mortar materials are 100 Bq kg^{-1} and 200 Bq kg^{-1}. When the ^{226}Ra activity concentration of a mortar material exceeds 200 Bq kg^{-1}, it is possible that radon exhalation from this material might cause indoor radon concentration to be higher than 200 Bq m^{-3} (EC, 2000). However, if the concentration is lower than 100 Bq kg^{-1}, then the resulting indoor radon concentration is going to be lower than 200 Bq m^{-3}. These considerations are reflected in the alpha index. The recommended limit concentration of ^{226}Ra is 200 Bq kg^{-1} for which I_α is equal to the unity (Rafique et al., 2011). Alpha index values are given in Table 3 and range from 0.15 to 0.24. The maximum value is observed in the basalt fine aggregate sample whereas the lowest value is found in cement (CEM I) sample. All observed values are less than the unity and it can be stated that the studied mortar materials are safe from considered environmental radiation hazards.

TABLE 4. Radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), gamma level index (I_γ) and alpha level index (I_α) for analyzed mortar material samples.

Sample Type	Ra_{eq} (Bq kg^{-1})	H_{ex}	H_{in}	I_γ	I_α
Cement (CEM I)	63.35	0.08	0.25	0.23	0.15
Silica fume	72.00	0.09	0.19	0.27	—
Fly ash	149.62	0.17	0.50	0.56	0.18
Limestone fine aggregate	—	—	—	—	—
Basalt fine aggregate	125.88	0.16	0.47	0.46	0.24

3.6 Calculation of Annual Effective Dose Rates

A conversion coefficient from the absorbed dose in the air to the effective dose 0.7 Sv Gy^{-1} and the indoor occupancy factor of 0.2 proposed by UNSCEAR (2000) was used in order to estimate the annual effective dose rates. Annual effective doses were calculated by the following relations (Eq. 6 and Eq. 7) (Shoeib and Thabayneh, 2014 and Rati et al., 2010).

$$\text{Indoor}\left(\text{mSv y}^{-1}\right) = \text{Absorbed dose}\left(\text{mGy h}^{-1}\right) \times 24\text{h} \times 365.25\text{d} \times 1.4 \times 0.8\left(\text{Sv Gy}^{-1}\right) \times 10^{-6} \quad [6]$$

And

$$\text{Outdoor}\left(\text{mSv y}^{-1}\right) = \text{Absorbed dose}\left(\text{mGy h}^{-1}\right) \times 24\text{h} \times 365.25\text{d} \times 0.2 \times 0.7\left(\text{Sv Gy}^{-1}\right) \times 10^{-6} \quad [7]$$

The calculated indoor, outdoor and total annual effective dose rates for studied mortar material samples are given in Table 5. It is obvious in the Table that the total annual effective dose values for each sample are less than the corresponding 1 mSv y^{-1} for a material to be used in the construction of dwellings. Hence, the studied mortar materials are safe to be used as mortar materials.

3.7 Presumption of the Absorbed Gamma Dose Rate

The indoor-absorbed dose rates (D) due to mortar materials were calculated from ^{226}Ra, ^{232}Th and ^{40}K concentration values included in those materials. The applied conversion factors used to compute D ($nGyh^{-1}$) are 0.462, 0.621 and 0.0417 for radium, thorium and potassium, respectively (El-Shershaby et al., 2006; Shoeib and Thabayneh, 2014).

$$D\left(\text{nGy h}^{-1}\right) = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_{K} \quad [8]$$

Table 5 shows that the absorbed dose rate values vary between 29.67 and 71.96 nGy h^{-1}. The absorbed dose rate value of limestone fine aggregate was not calculated due to the non-detectable ^{226}Ra activity concentration of limestone. The absorbed dose rate values are below the world average indoor absorbed gamma dose rate of 84 nGy h^{-1}.

TABLE 5. Indoor (D_{in}), outdoor (D_{out}) and total (D_{tot}) annual effective dose equivalent and absorbed dose rate (D) for studied mortar material samples.

Sample Type	D_{in} (mSv h^{-1})	D_{out} (mSv h^{-1})	D_{tot} (mSv h^{-1})	D(nGy h^{-1})
Cement (CEM I)	0.29	0.04	0.33	29.67
Silica fume	0.34	0.04	0.38	34.20
Fly ash	0.71	0.09	0.79	71.96
Limestone fine aggregate	—	—	—	—
Basalt fine aggregate	0.58	0.07	0.65	59.12

4. CONCLUSION

In this study, radon activity concentration, Radium (^{226}Ra), Thorium (^{232}Th) and Potassium (^{40}K) activity concentrations, Radium equivalent activity values, external and internal hazard indexes, radioactivity level index (gamma index), alpha index, annual effective dose rates and absorbed gamma dose rate were determined. The radon activity concentrations are ranked among the ones by ICRP, except for basalt. ^{226}Ra, ^{232}Th and ^{40}K concentration levels are in the range of international levels. It is generally observed that most of the data obtained in this study are regarded within the international range. It was determined that Ra_{eq} values of the construction material samples were within world standards. It is observed that limestone samples hold low radioactivity concentrations which were not measured. In Table 3, several literature data are represented, and it is noted that the lowest radionuclide activity concentration levels were measured in limestone samples. Obtained results demonstrate that measured raw materials collected from İzmir and its neighbourhood show low radiological impact in terms of radioactivity activity, dose levels and so on. It can be concluded from the overall results in this study that the studied mortar materials do not pose any significant source of radiation hazard.

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Appendix A

TABLE A1. Symbol and unit abbreviation

Symbol	Term
α	Alpha particle
γ	Gamma ray
R_{eq}	Radium equivalent activity
H_{ex}	External hazard index
H_{in}	Internal hazard index
I_{α}	Alpha level index
I_{γ}	Gamma level index
h	Hour
y	Year
Unit abbreviation	
Bq	Becquerel
Gy	Gray
Sv	Sievert
mSv	Millisievert
mSv/h	Millisievert per year
nGy/h	Nano Gray per hour

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