

# Radon exhalation and health hazard assessment for various ceramic tiles used in Bangladesh

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## Abstract

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**Objective:** Among various building materials, tiles are the most used decorative materials used worldwide. For the safe use of tiles at home and workplace it is required to select them properly based on radiation dose due to radon and gamma emission.

**Experiments:** Among various tiles available in the market, 25 different types of tiles were collected for this experiment. Collected tiles are then ground and stored in sealed cans for secular equilibrium. To measure gamma activity due to the radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$ , High Purity Germanium (HPGe) detector was used. To store radon gas emitting from tiles, a big box was used. The activity concentration was then measured by a radon detector (RadonEye) by placing it inside the box.

**Results and discussion:** It was observed that  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$  activity varies from 45 – 89 Bq/Kg, 77 – 110 Bq/Kg, and 321 – 694 Bq/Kg. The radium equivalent activity varies from 198.32 – 280.46 Bq/Kg, less than the recommended value of 370 Bq/Kg set by UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). The activity concentration of radon varies from 31 – 71 Bq/m<sup>3</sup>. This is lower than the 300 Bq/m<sup>3</sup> recommended by ICRP (International Commission on Radiological Protection). The radon exhalation rate varies from 0.179 – 0.409 Bqm<sup>-2</sup>h<sup>-1</sup>. To assess radiological hazards associated with the tiles samples air absorbed dose rate, internal and external hazard index, gamma index, annual indoor and outdoor effective dose rate, and dose due to radon were also calculated.

**Conclusion:** Though the estimated values for external and internal hazards are below the recommended values, they may be safe for adults. As children have thinner skin and breathe more air due to high breathing frequency, they may receive more radiation than adults. Moreover, they are at a greater risk because of their developing bodies and long life expectancy post-exposure.

**Keywords:** radionuclides; tiles; radon; tiles radiation; tiles health hazard; radon exhalation; tiles radon; radon cancer risk

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$^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  are the main sources of natural radiation that human beings are exposed to. There are two ways of radiation exposure, namely, external and internal. External health hazards are due to gamma radiation, and internal health hazards are for inhalation of radon and thoron gas emitted from Radium and Thorium, respectively. Depending on the distribution of these radionuclides, in building material, the dose varied in different ways. One of the most used building materials in modern houses is tiles which are made from sand, clay, and other natural materials. When it is required to make the glaze, zircon sand (Zirconium Silicate,  $\text{ZrSiO}_4$ ) is used because of the shining property of the lanthanum series. When impurities are added, zirconium silicate produces various colors. Deng et al. (1) found high concentrations of the radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in zircon sand. The activity concentration in zircon sand is 17,500 Bq kg<sup>-1</sup>, which is 106 times as much as that in ordinary building materials.

Activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in tiles varied in different ways depending on the materials used to make them. Table 2 shows activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  used in different countries' tiles. According to ICRP (2), people spend 80% of their time indoors and, hence, receive both internal and external doses throughout their lives. Depending on the building materials, sometimes the dose could be higher than the dose criteria set by ICRP or UNSCEAR (3). According to Papastefanou et al. (4), it could be five times higher than the recommended value when tiles are used as a building material.

Though the radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  emit gamma radiation, the first two are responsible for radon and thoron gas. According to the recent review of the International Agency for Research on Cancer (IARC), radon is one of the group-1 carcinogenic agents (5). Epidemiological studies confirmed that radon in homes increased the risk of lung cancer in the general

population. Due to these carcinogenic properties, people from different parts of the world paid attention to measuring doses due to radon inhalation and its exhalation rate. US Environmental Protection Agency (EPA) (6) estimated that out of a total of 157,400 lung cancer deaths nationally in 1995, 21,100 (13.4%) were radon related. The European studies reported in (7) showed the risk of lung cancer increased by 8% per 100 Bq/m<sup>3</sup> increase in measured radon concentration (95% confidence interval 3–16%). But this number is increased to 16% per 100 Bq/m<sup>3</sup> (95% confidence interval 5–31%) when the long-term average is measured considering a random year-to-year variability of radon concentration.

As thoron has the same properties as radon, it may also have the same carcinogenic effect on humans. However, due to the high price of the device, thoron measurement research is very limited. Though many researchers are doing their best to build radon thoron discriminative detectors (8–11). As the half-life of radon is much higher than that of thoron, it is believed that thoron has negligible health hazards than radon. By reducing the radon concentration at home below 4 picocuries per liter (pCi/L), lung cancer patient number could be reduced to one-fourth (6).

To determine the population's exposure to indoor radiation, it is required to measure radiation dose due to building materials. It is also required to set national guidelines and standards for the safe use of these types of building materials. The main objective of this project is to calculate the gamma activity concentration, radon exhalation rate, and associated hazard indices for natural radionuclides present in the tiles made in Bangladesh and imported from abroad. So that people can decide which tiles will be suitable for their home and workplace. At the same time, it may be helpful to the tile companies in selecting the materials for their product.

## Materials and methods

For this research, 25 different tiles made by different companies were collected from the local market. Among 25 tiles samples, 16 were made in Bangladesh, one was made in Germany, one was from UAE, and seven others were from China. After collection, the tiles were washed with ethanol to remove external contamination, and then they were ground with a grinder. After grinding each sample, the grinder was cleaned by using a high-pressure blower to avoid cross-contamination of the samples. After that, they were dried in an oven for 5 days at 100°C.

### Gamma analysis

As the energy resolution of the high purity germanium (HPGe) detector is high, the measurement using the detector is non-destructive, and the information provided by it is accurate and reliable, we have used HPGe for radioactivity analysis of the samples through gamma emission. For

gamma analysis, 500 g of sieved and dried samples was weighed out with Marinelli beakers with a height of 10 cm and an inner diameter of 6 cm. After sealing the beakers, they were kept in the laboratory for 2 months to obtain a nearly secular equilibrium between <sup>226</sup>Ra and its shorter-lived daughter products. The analysis was carried out using the gamma-ray spectrometry facilities at the Atomic Energy Centre, Chattogram, Bangladesh. The NATS-2 2465-17 HPGe detector with a relative efficiency of 40% at 1.33 MeV photon with respect to 3"×3" NaI detector and Co-60 source mounted 25 cm above the detector. Efficiency calibration of the detector was carried out by using standard soil-327 from the International Atomic Energy Agency (IAEA). Canberra Genie-2000 software was used to analyze the spectrum. After efficiency calibration, the tiles samples are counted for 30,000 sec in the HPGe detector system.

### Radon exhalation

To measure the exhalation rate of radon, at first, the tiles powder was spread on a tray (0.371 m<sup>2</sup>) that was placed at the bottom of the airtight box with 0.283 m<sup>3</sup> volumes. A RadonEye detector was then placed on the stand inside the box. To decay out the background radon (those that enter into the box during sample changing time) and to allow the gas to reach a saturation level, the box was closed for 2 weeks before taking any readings. The radon gas collection system is shown in Fig. 1.

Inside a chamber, the radon accumulation rate ( $C_{Rn}^a$ ) can be expressed as a function of time by the following the equation (12):

$$\frac{dC_{Rn}^a}{dt} = \frac{Exh.A}{V_c} + C_{Rn}^B \lambda_w - \lambda_{eff} C_{Rn}^a \quad (1)$$

where ( $c_{Rn}^b$ ) is the background concentration of radon in the chamber.  $V_c$  is the free volume of the closed chamber.  $Exh$  is the exhalation rate, and  $A$  is the surface area of the sample.  $\lambda_{eff}$  = effective radon removal rate.  $\lambda_w$  is the leakage rate.  $\lambda_{eff} = \lambda_{Rn} + \lambda_{bd} + \lambda_w$ , where  $\lambda_{bd}$  is the radon back

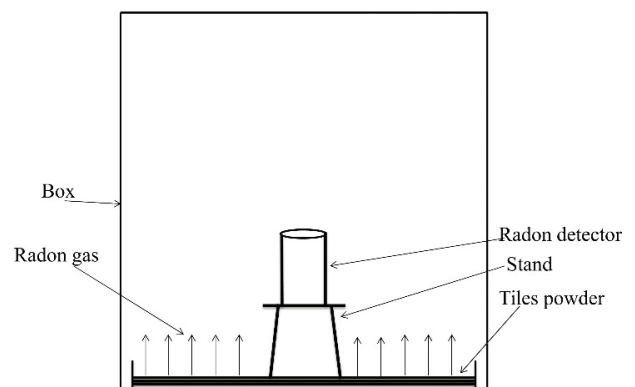


Fig. 1. Schematic diagram of radon gas collection system.

diffusion rate.  $\lambda_{Rn}$  is the decay constant of radon. Solution of equation (1) with boundary condition  $C_{Rn}^a(0) = C_{Rn}^0$  becomes

$$C_{Rn}^a(t) = C_{Rn}^0 e^{\lambda_{eff} t} + \left( \frac{Exh.A}{\lambda_{eff} \cdot V_C} + \frac{C_{Rn}^B \lambda_B}{\lambda_{eff}} \right) (1 - e^{-\lambda_{eff} \cdot t}) \quad (2)$$

For a tight chamber, there is no leakage of radon, and as the volume of the chamber is much bigger than 1 L, the aforementioned equation for a saturated radon condition can be written as

$$Exh = \frac{C_{Rn}^{Sat} \lambda_{Rn} V_C}{A} \quad (3)$$

where  $C_{Rn}^{Sat}$  is radon concentration at saturation condition.

#### Radiological hazards

According to the formula reported in (3), the indoor air absorbed dose rate at 1 m above the ground surface is calculated by the following equation. As the dose due to other radionuclides like  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{235}\text{U}$ , etc. decay series is very low compared to the total dose from an environmental background, we can neglect them.

$$D_{Air} = (0.461A_{Ra} + 0.623A_{Th} + 0.0414A_k) nGy h^{-1} \quad (4)$$

where  $A_{Ra}$ ,  $A_{Th}$ , and  $A_k$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  of the samples.

The levels of radionuclides  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in the tiles are not uniformly distributed. For this reason, to compare the radioactivity of each radionuclide, radium equivalent activity was used. In this case, it is assumed that 370 Bq/kg  $^{226}\text{Ra}$ , 259 Bq/kg  $^{232}\text{Th}$ , or 4,810 Bq/kg  $^{40}\text{K}$  produces the same gamma dose rate. The radium equivalent activities are calculated by the following equation (13):

$$Ra_{eq} = A_{Ra} + (1.43 \times A_{Th}) + (0.07 \times A_k) \quad (5)$$

where  $A_{Ra}$ ,  $A_{Th}$ , and  $A_k$  are the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  of the samples, respectively.

The external hazard index  $H_{ex}$  was calculated by using the following formula (14):

$$H_{ex} = \left( \frac{A_{Ra}}{370} \right) + \left( \frac{A_{Th}}{259} \right) + \left( \frac{A_K}{4810} \right) \quad (6)$$

The internal hazard index was calculated by the following equation:

$$H_{in} = \left( \frac{A_{Ra}}{185} \right) + \left( \frac{A_{Th}}{259} \right) + \left( \frac{A_K}{4810} \right) \quad (7)$$

To calculate the annual effective dose rate for indoors, the following equation was used:

$$AEDR_{indoor} (mSv) = D_{Air} (nGy h^{-1}) \times 24 \times 365 (h^{-1}) \times 0.8 \times 0.7 (Sv \cdot Gy^{-1}) \times 10^{-6} \quad (8)$$

where 0.8 is the indoor occupancy factor, which implies that people spend 20% of the time outdoors on average, around the world. The coefficient is 0.7 SvGy<sup>-1</sup> to convert the absorbed dose in the air to an effective dose equivalent and effective dose (3).

The gamma index is used to calculate the  $\gamma$  radiation hazard related to the natural radionuclide in the particular samples under investigation. The gamma index representation ( $I_\gamma$ ) is estimated by the following equation, and the unit of this is Bq/Kg.

$$I_\gamma = \left( \frac{A_{Ra}}{300} \right) + \left( \frac{A_{Th}}{200} \right) + \left( \frac{A_K}{3000} \right) \quad (9)$$

The annual effective dose due to radon can be calculated by the following equation suggested by (15):

$$D_{Rn} (mSv \times y^{-1}) = C_{Rn} \cdot T \cdot f_{Rn} \cdot F_{eq} \quad (10)$$

where  $f_{Rn}$  is the conversion factor =  $9 nSv (Bq \cdot h \cdot m^{-3})^{-1}$ . T is the time spent indoors per year = 7,008 h as the indoor occupancy factor is 80% of 8,760 h per year.  $F_{eq}$  is the equilibrium fraction, and for indoor, this value is 0.4.

Substituting the values to the aforementioned equation gives an indoor dose

$$D_{Rn} (mSv \times y^{-1}) = 0.02523 \times C_{Rn} \quad (11)$$

## Result and discussion

In this experiment, a total of 25 tiles samples were analyzed to measure dose due to gamma radiation and radon inhalation. The radon exhalation rate was measured by using a sealed box system for every sample, and activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  were measured by using the HPGe detector. Table 1 shows the activity concentration of the aforementioned radionuclide in the most used tiles in Bangladesh.

We found a moderate correlation between  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  with an  $R^2$  value of 0.326, as shown in Fig. 2.

It was also observed that tiles used in different countries have different activity concentrations even though the country of origin is the same. Table 2 shows the results of different experiments on tile radiation conducted in different countries.

The hazard index due to tiles used in the present study is shown in Table 3.

Radon exhalation rate and dose due to inhalation have shown in Table 4.

To be acceptable, the external radiation hazard  $H_{ex}$  must be less than unity (15). In this experiment, we found this value varies from 0.543 to 0.767. Like  $H_{ex}$ , the

internal hazard index  $H_{in}$  must be less than unity to be acceptable. Here, it varies from 0.665 to 1.007.

The gamma activity index  $\leq 1$  corresponds to an annual effective dose less than or equal to  $1 \text{ mSv} \cdot \text{y}^{-1}$ , while the gamma activity index  $\leq 0.5$  corresponds to  $0.3 \text{ mSv} \cdot \text{y}^{-1}$  if the materials are used in bulk quantity. In this research, the gamma index of the imported tiles varies from 0.774 to 1.009, whereas this value for local tiles varies from 0.723 to 0.999.

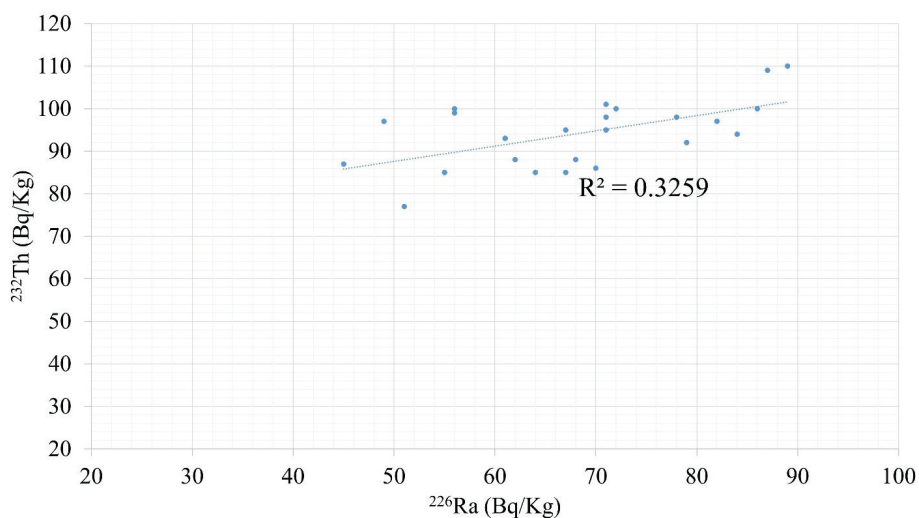
It was observed that the radon concentration is positively correlated with  $^{226}\text{Ra}$  concentration in the tiles with an  $R^2$  value of 0.786, as shown in Fig. 3.

**Table 1.** The activity concentration of the  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$

Origin	Sample code	$^{226}\text{Ra}$ (Bq/Kg)	$^{232}\text{Th}$ (Bq/Kg)	$^{40}\text{K}$ (Bq/Kg)
Bangladesh	BD-1	56 ± 4.2	99 ± 7.4	580 ± 63.7
Bangladesh	BD-2	72 ± 5.9	100 ± 7.4	550 ± 61.3
Bangladesh	BD-3	87 ± 6.4	109 ± 7.6	492 ± 56.8
Bangladesh	BD-4	70 ± 5.7	86 ± 6.4	528 ± 58.5
Bangladesh	BD-5	51 ± 3.8	77 ± 5.9	601 ± 66.1
Bangladesh	BD-6	64 ± 4.6	85 ± 6.3	694 ± 72.9
Bangladesh	BD-7	56 ± 4.2	100 ± 7.4	498 ± 57.2
Bangladesh	BD-8	61 ± 4.4	93 ± 6.8	602 ± 66.5
Bangladesh	BD-9	71 ± 5.7	95 ± 7.2	556 ± 62.1
Bangladesh	BD-10	61 ± 4.4	93 ± 6.8	608 ± 66.8
Bangladesh	BD-11	45 ± 3.2	87 ± 6.4	413 ± 52.6
Bangladesh	BD-12	49 ± 3.5	97 ± 7.3	436 ± 53.4
Bangladesh	BD-13	62 ± 4.4	88 ± 6.5	485 ± 55.7
Bangladesh	BD-14	82 ± 6.3	97 ± 7.3	321 ± 28.8
Bangladesh	BD-15	79 ± 6.1	92 ± 6.8	463 ± 54.1
Bangladesh	BD-16	68 ± 5.5	88 ± 6.5	487 ± 55.8
UAE	UAE-1	71 ± 5.7	98 ± 7.4	521 ± 58.2
Germany	Germany-1	86 ± 6.6	100 ± 7.5	527 ± 58.5
China	China-1	78 ± 6.1	98 ± 7.4	458 ± 53.9
China	China-2	55 ± 4.2	85 ± 6.3	498 ± 56.1
China	China-3	71 ± 5.7	101 ± 7.5	511 ± 57.4
China	China-4	67 ± 5.5	95 ± 7.2	521 ± 58.2
China	China-5	84 ± 6.4	94 ± 7.1	538 ± 59.3
China	China-6	89 ± 6.8	110 ± 8.0	488 ± 55.8
China	China-7	67 ± 5.5	85 ± 6.3	522 ± 58.2

**Table 2.** Comparative analysis of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  activity concentration in various tiles

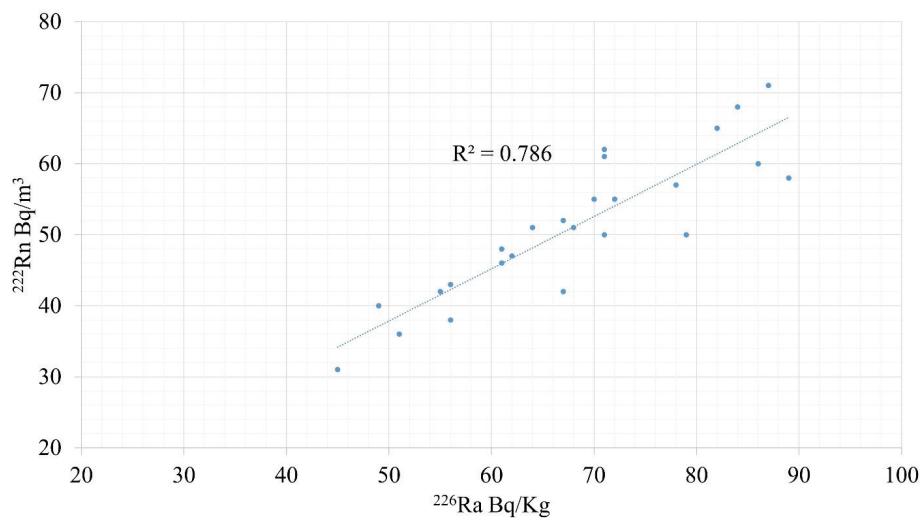
Country	$^{226}\text{Ra}$ BqKg <sup>-1</sup>	$^{232}\text{Th}$ BqKg <sup>-1</sup>	$^{40}\text{K}$ BqKg <sup>-1</sup>	References
China	63.5–131.4	55.4–106.5	386.7–866.8	(16)
China	42.5–75	41.5–405	290–740	(17)
Nigeria	27–241	41–461	270–860	(17)
Turkey	28.7–49.36	29.8–68.4	244.42–586.01	(18)
Turkey	54.2–105.1	44.8–88.2	325.7–1043.7	(19)
India	3.2–151.7	14–63.7	24.3–21.5	(20)
Italy	20–708	33–145	158–850	(21)
South Korea	44–82	34–96	310–1,019	(22)
Egypt	61–118	55–98	730–1,050	(23)
Greece	25–174	29–47	411–786	(24)
Sudan	93–318	16–239	126–384	(25)
EU	53–116	31–66	390–710	(26)
Croatia	16–82	7–42	71–430	(26)
Italy	17–252	20–94	60–1,000	(26)
Spain	37–111	30–77	188–1,110	(26)
Bangladesh	45–87	77–109	321–694	Present study
UAE	71	98	521	
Germany	86	100	527	
China	55–89	85–110	458–538	



**Fig. 2.** Correlation curve between the activity concentration of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ .

Table 3. Hazard index for the tiles

Tiles name	Ra <sub>eq</sub> (Bq/Kg)	D <sub>Air</sub> (nGyh <sup>-1</sup> )	H <sub>ex</sub>	H <sub>in</sub>	I <sub>γ</sub>	AEDR <sub>indoor</sub> (mSvy <sup>-1</sup> )
BD-1	238.170	110.028	0.654	0.806	0.875	0.540
BD-2	253.500	116.764	0.695	0.890	0.923	0.573
BD-3	277.310	126.694	0.758	0.993	0.999	0.622
BD-4	229.940	106.46	0.631	0.820	0.839	0.522
BD-5	203.180	95.312	0.560	0.698	0.755	0.468
BD-6	234.130	110.056	0.645	0.818	0.870	0.540
BD-7	233.860	107.188	0.641	0.792	0.853	0.526
BD-8	236.130	109.638	0.649	0.814	0.869	0.538
BD-9	245.770	113.534	0.674	0.866	0.897	0.557
BD-10	236.550	109.89	0.650	0.815	0.871	0.539
BD-11	198.320	90.684	0.543	0.665	0.723	0.445
BD-12	218.230	99.538	0.598	0.730	0.794	0.488
BD-13	221.790	102.166	0.608	0.776	0.808	0.501
BD-14	243.180	109.954	0.663	0.884	0.865	0.539
BD-15	242.970	111.512	0.665	0.878	0.878	0.547
BD-16	227.930	105.022	0.625	0.809	0.829	0.515
UAE-1	247.610	113.876	0.679	0.870	0.900	0.559
Germany-1	265.890	122.266	0.728	0.961	0.962	0.600
China-1	250.200	114.464	0.684	0.895	0.903	0.562
China-2	211.410	97.666	0.580	0.729	0.774	0.479
China-3	251.200	115.268	0.688	0.880	0.912	0.565
China-4	239.320	110.216	0.656	0.837	0.872	0.541
China-5	256.080	118.18	0.702	0.929	0.929	0.580
China-6	280.460	128.054	0.767	1.007	1.009	0.628
China-7	225.090	104.218	0.618	0.799	0.822	0.511

Fig. 3. Correlation between  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$ .

### Conclusion

In this research, we have measured natural radioactivity concentrations and calculated the associated radiological risk of 25 types of different tile samples collected from the local market. The average concentration of  $^{226}\text{Ra}$  for tiles

made in Bangladesh is 64.625 Bq/Kg, while for imported tiles, it is 73 Bq/Kg. For  $^{232}\text{Th}$ , the average value for Bangladeshi tiles is 92.875 Bq/Kg, while the value for China tiles is 95.42 Bq/Kg. For  $^{40}\text{K}$ , the average concentration is 476.62 Bq/Kg, while the value for imported one

**Table 4.** Radon exhalation rate and dose due to inhalation

Tiles name	<sup>226</sup> Ra (Bq/Kg)	C <sub>Rn</sub> (Bq/m <sup>3</sup> )	Exhalation rate (Bqm <sup>-2</sup> h <sup>-1</sup> )	D <sub>Rn (Indoor)</sub> (mSv/y)
BD-1	56	43	0.248	1.085
BD-2	72	55	0.317	1.388
BD-3	87	71	0.409	1.791
BD-4	70	55	0.317	1.388
BD-5	51	36	0.207	0.908
BD-6	64	51	0.294	1.287
BD-7	56	38	0.219	0.959
BD-8	61	46	0.265	1.161
BD-9	71	61	0.351	1.539
BD-10	61	48	0.276	1.211
BD-11	45	31	0.179	0.782
BD-12	49	40	0.230	1.009
BD-13	62	47	0.271	1.186
BD-14	82	65	0.374	1.640
BD-15	79	50	0.288	1.261
BD-16	68	51	0.294	1.287
UAE-1	71	62	0.357	1.564
Germany-1	86	60	0.346	1.514
China-1	78	57	0.328	1.438
China-2	55	42	0.242	1.060
China-3	71	50	0.288	1.261
China-4	67	42	0.242	1.060
China-5	84	68	0.392	1.716
China-6	89	58	0.334	1.463
China-7	67	52	0.300	1.312

is 644.55. The average value of radium equivalent activity of both imported and Bangladeshi tiles is below the recommended value of 370 Bq/Kg set by the UNSCEAR.

Different tiles possess different amounts of radiological hazards. This variation may be due to the variation of the material used to make the tiles attractive and strong. As ceramic tiles could be an extra source of radiation that contributes to the total annual dose reaching a higher level, manufacturers of the tiles should maintain the concentrations of radionuclides in their products to reduce health hazards.

### Conflict of interest and funding

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