

# Assessment of the long-term possible radiological risk from the use of ceramic tiles in Malaysia

Shittu Abdullahi<sup>2,3</sup> · Aznan Fazli Ismail<sup>1,2</sup> · Syazwani Mohd Fadzil<sup>1,2</sup> · Supian Samat<sup>2</sup>

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**Abstract** This study investigated the level of natural radioactivity and radiological risks of 40 different ceramic tiles through gamma-ray spectroscopy using a high-purity germanium detector. The calculated activity concentrations were evaluated to determine their potential radiological risks to human health. Furthermore, the activity concentrations were subjected to the RESRAD-BUILD computer code to assess the effect of ventilation rate, dweller position, and room size and direction on the total effective dose (TED). The simulated TED received by a receptor when changing the ventilation rate in a room ranged from  $0.26 \pm 0.01$  to  $0.61 \pm 0.01$  mSv/y; however, the percentage variations in the TED due to dweller position and room size are 34, 31, and 35% and 33, 27, and 40% for the x-, y-, and z-directions, respectively. The overall TED received by the dweller based on room size and direction is 0.75 mSv/y. The calculated radiological risk parameters were all below the recommended maximum limit. However, the TED received by the dweller is significantly affected by the directions of the measurement, position, room size, and

ventilation. Therefore, estimating the TED from one direction would underestimate the total dose received by the dweller.

**Keywords** Radiological risk · RESRAD-BUILD computer code · Ceramic tile · Room size · Ventilation rate

## 1 Introduction

All building materials contain numerous radionuclides [1, 2]. Materials originating from rock and soil contain uranium ( $^{238}\text{U}$ ), thorium ( $^{232}\text{Th}$ ), and their decay series and singly occurring potassium ( $^{40}\text{K}$ ) [3]. These radionuclides undergo continuous natural decay that subsequently releases radiation to the environment. Radium ( $^{226}\text{Ra}$ ) is the reference and most significant radionuclide in the measurement of  $^{238}\text{U}$ . Natural radiation contributes to over 80% of the total exposure received by the general population. The aforementioned radionuclides and their progenies are the most common radionuclides found in building materials. Continuous exposure to these radionuclides from building materials via their decay chains and singly occurring  $^{40}\text{K}$  may pose a radiological risk to dwellers [2].

Ceramic tiles are common indoor decorative materials and are made from natural products that have been industrially processed through pressing into shapes and firing at a high temperature. The body of the tile may be glazed or unglazed, depending on its purpose. Most ceramic tiles have the following properties: water absorption level ranging from 0.5 to 10% [4] and high mechanical and chemical characteristics. The manufacturing of ceramic tiles includes the addition of a chemical agent, such as zircon, to act as an opacifier [2, 4, 5]. In this regard, these

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✉ Aznan Fazli Ismail  
aznan@ukm.edu.my

<sup>1</sup> Nuclear Science Program, Faculty of Science and Technology, Universiti Kebangsaan Malaysia (UKM), 43600 Bangi, Selangor, Malaysia

<sup>2</sup> Centre for Frontier Science, Faculty of Science and Technology, Universiti Kebangsaan Malaysia (UKM), 43600 Bangi, Selangor, Malaysia

<sup>3</sup> Department of Physics, Faculty of Science, Gombe State University, P.M.B. 127, Gombe, Nigeria

tiles may have natural activity concentrations higher than the recommended maximum limit and contribute to the indoor radiation dose received by the dwellers. Therefore, it is necessary to research and evaluate the level of natural radioactivity and potential radiological hazards posed by tile materials.

The aim of this study is to determine the level of natural radioactivity from ceramic tiles and evaluate the calculated activity concentrations that pose potential radiological risks to human health. Furthermore, the activity concentrations are subjected to the RESRAD-BUILD computer code to investigate the effects of ventilation rate, dweller position, and room size and direction on the total effective dose received by the dweller.

## 2 Materials and methods

### 2.1 Sample preparation

The studied materials are common ceramic tiles used for flooring, structuring, and decorating indoor and outdoor rooms in Malaysia. Forty sample ceramic tiles were purchased from local suppliers in Kajang, Johor, Pahang, Terengganu, Kelantan, Kuala Lumpur, and Putrajaya. The samples were dried in an oven for 48 h at 105 °C, ground to obtain a homogeneous powder, and sieved through a 500- $\mu$ m mesh to ensure uniform particle sizes. The prepared samples were then packed in a counting bottle and stored in the laboratory for 30 days to reach the secular equilibrium. Three replicates of each sample were prepared, as proposed by IAEA technical report series number 259 [3, 6].

### 2.2 Analysis of natural radioactivity

The samples were analyzed through gamma-ray spectroscopy with a high-purity germanium (HPGe) detector connected to a personal computer analyzer (PCA) with Genie 2000 software. The HPGe detector is enclosed within a 10-cm-thick lead CANBERRA 747 shielding coated with 1 and 1.6 mm of tin and copper, respectively, to minimize the interference of background radiation and cosmic rays in the detector environment. A mixture of  $^{22}\text{Na}$ ,  $^{57}\text{Co}$ ,  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$  standards was used for energy calibration. The energy photo-peaks used to determine the activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  in the detector were 1764 ( $^{214}\text{Bi}$ ), 2614 ( $^{208}\text{Tl}$ ), and 1461 keV ( $^{40}\text{K}$ ), with equivalent emission probabilities of 15.2, 99.8, and 10.7%, respectively [8]. Each sample, including the background study, was counted using a preset time of 12 h.

The performance criterion of gamma-ray spectroscopy was characterized using parameters such as efficiency,

critical level ( $L_c$ ), lower limit of detection ( $LLD$ ), and minimum detectable activity ( $MDA$ ). The  $LLD$  is the level at which a true net count is detected above the critical level ( $L_c$ ) with a given probability when actual activity occurs. Meanwhile, the  $MDA$  is the lowest number of detected net counts through a certain degree of confidence and depends on the  $LLD$  and efficiency of a counting system which was previously calculated. The background counts play a significant role in both the  $LLD$  and  $MDA$ . The aforementioned parameters are calculated using Eqs. 1, 2, and 3 [8, 9], and the results are presented in Table 1.

$$L_c = 2.33\sqrt{N_B}, \quad (1)$$

$$LLD = 2.71 + 4.65\sqrt{N_B}, \quad (2)$$

$$MDA (\text{Bq kg}^{-1}) = \frac{LLD}{t\varepsilon PM}, \quad (3)$$

where  $N_B$  is the background count and  $t$ ,  $\varepsilon$ ,  $P$ , and  $M$  are the counting time, efficiency at the energy of interest, emission probability, and mass of the sample in kg, respectively.

## 3 Results and discussion

### 3.1 Activity concentration of $^{226}\text{Ra}$ , $^{232}\text{Th}$ , and $^{40}\text{K}$

The activity concentrations of individual radionuclides are calculated using Eq. (4)

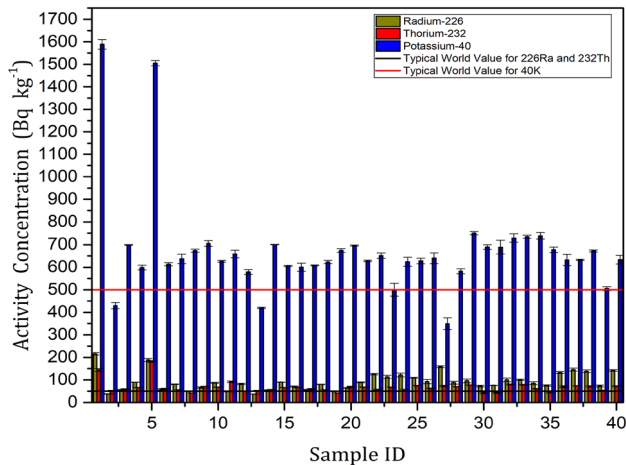
$$C = \frac{m_{\text{std}} \times A_s}{m_s \times A_{\text{std}}} C_{\text{std}}, \quad (4)$$

where  $C$  and  $C_{\text{std}}$  are the activity concentration of individual radionuclides and standard soil, respectively (in Bq/kg). In this case, IAEA-SRM soil-375 was used as a standard reference material.  $m_s$  and  $m_{\text{std}}$  are the mass of the sample and standard material (kg), while  $A_s$  and  $A_{\text{std}}$  are the net count of the sample and standard material in count per second (cps), respectively,

The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  for the tile materials are shown in Fig. 1. The distribution of the aforementioned radionuclides is non-uniform across the various tile materials. The activity concentrations range from  $39 \pm 0.3$  to  $216 \pm 6$  Bq/kg,  $42 \pm 0.1$  to  $182 \pm 4$  Bq/kg, and  $350 \pm 30$  to  $1589 \pm 20$  Bq/kg for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , respectively, with mean values of  $93 \pm 40$ ,  $68 \pm 30$ , and  $673 \pm 200$  Bq/kg, respectively. The United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) [11] reported activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  for soil in Malaysia ranging from 38 to 94 Bq/kg, 63 to 110 Bq/kg, and 170 to 430 Bq/kg, with mean values of 67, 82, and 310 Bq/kg, respectively. Hence, the reported activity concentrations of the three radionuclides are comparable to those of

**Table 1** Critical level ( $L_c$ ), lower limit of detection ( $LLD$ ), and minimum detectable activity ( $MDA$ ) for the counting system

Radionuclides	Gamma-ray energies (keV)	$L_c$ (counts)	$LLD$ (counts)	$MDA$ (Bq/kg)
$^{40}\text{K}$	1461	$55 \pm 0.8$	$112 \pm 2$	$22 \pm 0.2$
$^{214}\text{Bi}$	1764.5	$32 \pm 0.3$	$67 \pm 0.5$	$6 \pm 0.1$
$^{208}\text{Tl}$	2614.5	$42 \pm 2$	$86 \pm 4$	$5 \pm 0.1$

**Fig. 1** (Color online) Activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$ , and typical world value

Malaysian soils. However, the mean values reported here are mostly higher than the activity concentrations in Malaysia's soil [11].

To validate the current findings, this study was compared to similar studies conducted around the world, as given in Table 2. The activity concentrations of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  reported here are comparable to those in almost all other studies, excluding those reported from Egypt and Italy, which are slightly lower than those in other studies. However, the activity concentration of  $^{40}\text{K}$  reported here is comparable to that of studies conducted in China, Egypt,

and Korea, while higher than those reported from Italy and Yemen.

### 3.2 Evaluation of Radiological Hazard Index

The radiological hazard index was evaluated using the radium equivalent activity ( $Ra_{eq}$ ) determined by Eq. (5) and activity concentration index ( $ACI$ ) determined by Eq. (6) [12–16]:

$$Ra_{eq}(\text{Bq/kg}) = C_{Ra} + 1.43C_{Th} + 0.077C_K, \quad (5)$$

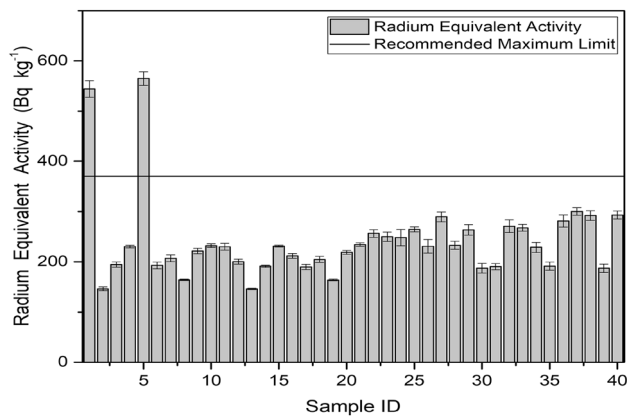
$$ACI = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000}. \quad (6)$$

The radium equivalent activity is used to assess the potential radiological risk attributed to the studied tile materials. The activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  are non-uniformly distributed among different building materials and even among materials of the same category. Beretka and Mathew [17] reported that the expression of radium equivalent activity was derived based on the assumption that 370, 259, and 4810 Bq/kg of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , produced the same gamma dose rate [17]. Therefore, the radium equivalent activity was calculated from the activity concentration of the three aforementioned radionuclides using Eq. (2) [9, 16, 17]. The evaluated radium equivalent activity is presented in Fig. 2, with mean values ranging from  $146 \pm 1$  to  $565 \pm 10$  Bq/kg. The radium equivalent activity results reported here were all below the recommended maximum limit of 370 Bq/kg,

**Table 2** Comparison of this study and others from various parts of the world

Country	$^{226}\text{Ra}$ (Bq/kg)	$^{232}\text{Th}$ (Bq/kg)	$^{40}\text{K}$ (Bq/kg)	$Ra_{eq}$ (Bq/kg)	Sources
Anhui, China	$52 \pm 8$	$69 \pm 10$	$746 \pm 50$	207	[18]
Egypt	$60 \pm 30$	$47 \pm 20$	$703 \pm 200$	$181 \pm 60$	[19]
Korea	64	75	677	*224	[20]
Bangladesh	$61 \pm 2$	$71 \pm 2$	$1000 \pm 10$	239	[1]
Yemen	132	84	401	308	[21]
Italy	33	32	436	208	[22]
Japan	83	64	527	*215	[23]
India	42	57	528	165	[24]
Nigeria	$61 \pm 6$	$70 \pm 6$	$515 \pm 60$	$204 \pm 40$	[25]
TWAV	50	50	500	370	[26]
Malaysia	$92 \pm 40$	$68 \pm 30$	$673 \pm 200$	$241 \pm 80$	Present study

The values marked with \* were not computed by the respective authors, but were computed here for comparison purposes. TWAV is the typical world average value



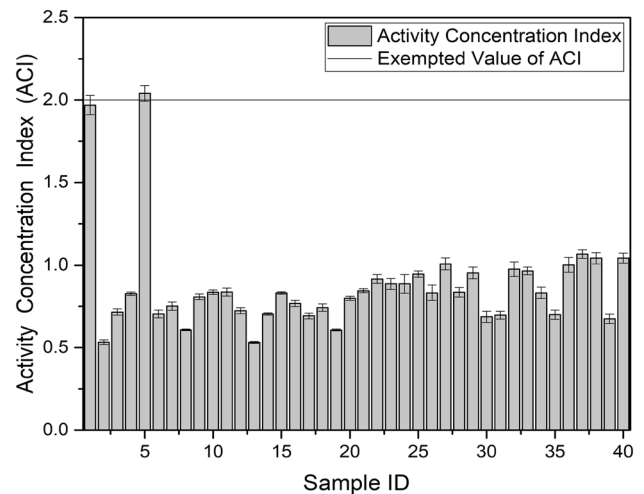
**Fig. 2** Radium equivalent activity and recommended value

except for those of tiles 1 and 5. The radium equivalent activities ( $Ra_{eq}$ ) reported here were also compared to those from other studies conducted around the world and are presented in Table 1. The comparison showed that the values reported here are analogous with those of all other compared studies.

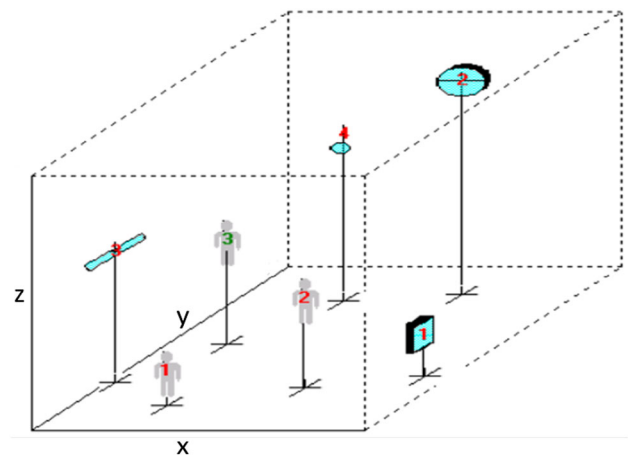
The  $ACI$ , also known as the gamma index, was proposed by the European Commission (EC). The purpose of this index is to verify whether the EC's guidelines for the usage of building materials are met. The EC recommends that the used building materials should meet the control guidelines based on a dosage range of 0.3 to 1 mSv/y. This is the gamma dosage surplus to that received outdoors if an exemption limit of 0.3 mSv/y is adopted. Therefore, the  $ACI$  value should be below 0.5 for building materials such as brick, sand, gravel, and cement, which are used in bulk. However, the  $ACI$  value should be below 2 for superficial building materials such as tiles and boards, which are used in restricted quantities. If the upper limit of 1 mSv/y is adopted, the  $ACI$  value should be below 1 and 6 for bulk and restricted building materials, respectively [15, 16]. The computed  $ACI$  is presented in Fig. 3, with mean values ranging from  $0.5 \pm 0.01$  to  $2.0 \pm 0.05$ . The values reported here were all below the exemption limit, except for tile 5, which presented a slightly higher value than the exemption limit, but lower than the upper limit.

### 3.3 Radiological Dose Assessment Using the RESRAD-BUILD Computer Code

The RESRAD-BUILD computer code designed by Argonne National Laboratory is a pathway analysis model designed to assess the possible radiological dose incurred by a dweller living or working in a building contaminated with radioactive materials, which can be released into the indoor air by the mechanisms such as diffusion, mechanical removal, or erosion [27, 28].



**Fig. 3** Activity concentration index and exempted limit



**Fig. 4** (Color online) Three-dimensional view of the room description [27]

The radiological dose was assessed in this study by varying the room size from 6 to 42 m<sup>2</sup>. The dose library from the International Commission on Radiological Protection (ICRP 72) was adopted, with exposure times ranging from 0 to 70 y [27]. The breathing rate, time fraction, and ingestion rate are 20 m<sup>3</sup>/d, 0.8, and 0 m<sup>2</sup>/h, respectively. The default code parameters, such as deposition velocity, resuspension rate, emanation power, and air fraction, were used for other inputs. Other essential parameter determined is the position of the receptor which is assumed to be at the center of the room, while the dimensions of the room are width  $\times$  length  $\times$  height, denoted by  $x$ ,  $y$ , and  $z$ , respectively.

**Table 3** Room parameters and dweller position

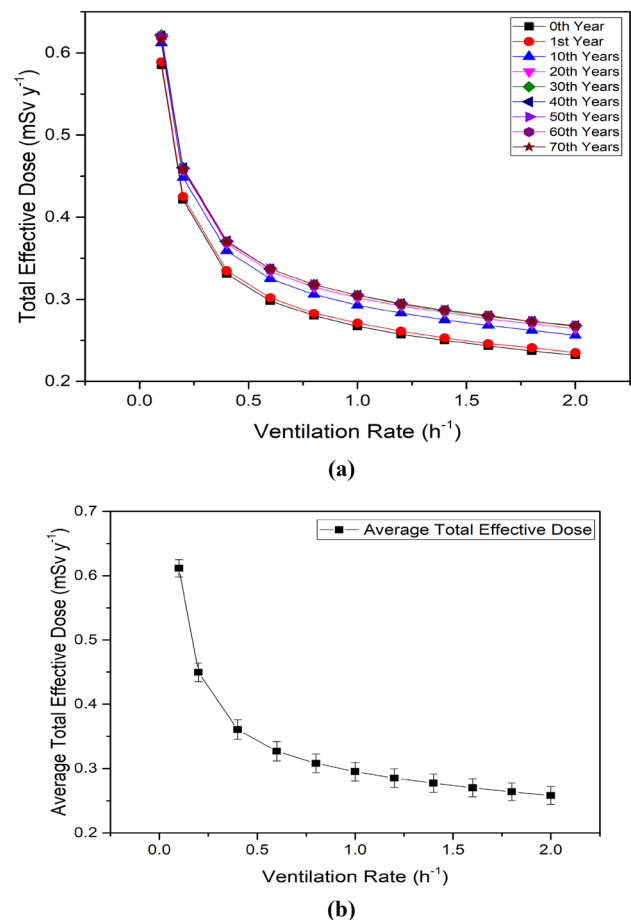
Room size (width $\times$ length $\times$ height) m <sup>3</sup>	Position of the dweller	Room area (m <sup>2</sup> )
2 $\times$ 3 $\times$ 3	1 $\times$ 1.5 $\times$ 1	6
3 $\times$ 3 $\times$ 3	1.5 $\times$ 1.5 $\times$ 1	9
3 $\times$ 4 $\times$ 3	1.5 $\times$ 2 $\times$ 1	12
3 $\times$ 5 $\times$ 3	1.5 $\times$ 2.5 $\times$ 1	15
4 $\times$ 5 $\times$ 3	2 $\times$ 2.5 $\times$ 1	20
5 $\times$ 5 $\times$ 3	2.5 $\times$ 2.5 $\times$ 1	25
5 $\times$ 6 $\times$ 3	2.5 $\times$ 3 $\times$ 1	30
6 $\times$ 6 $\times$ 3	3 $\times$ 3 $\times$ 1	36
6 $\times$ 7 $\times$ 3	3 $\times$ 3.5 $\times$ 1	42

### 3.3.1 Room Description Used by the RESRAD-BUILD Computer Code

A coordinate system is used to define the location of the contaminated sources and dweller points inside the building. The RESRAD-BUILD code is flexible and allows the origin and coordinates axes to be placed at any location and direction by the user. However, the origin is normally located at the bottommost left corner, as shown in Fig. 4, where the  $x$ -axis measures the horizontal distance (width of the room or short wall) to the right of the origin and the  $y$ -axis measures the length (long wall) of the room perpendicular to the  $x$ -axis. The  $z$ -axis measures the height (the vertical distance) of the room. The room parameters and dweller positions are presented in Table 3.

### 3.3.2 Long-term Effect of Ventilation on the Total Effective Dose Received by Dweller

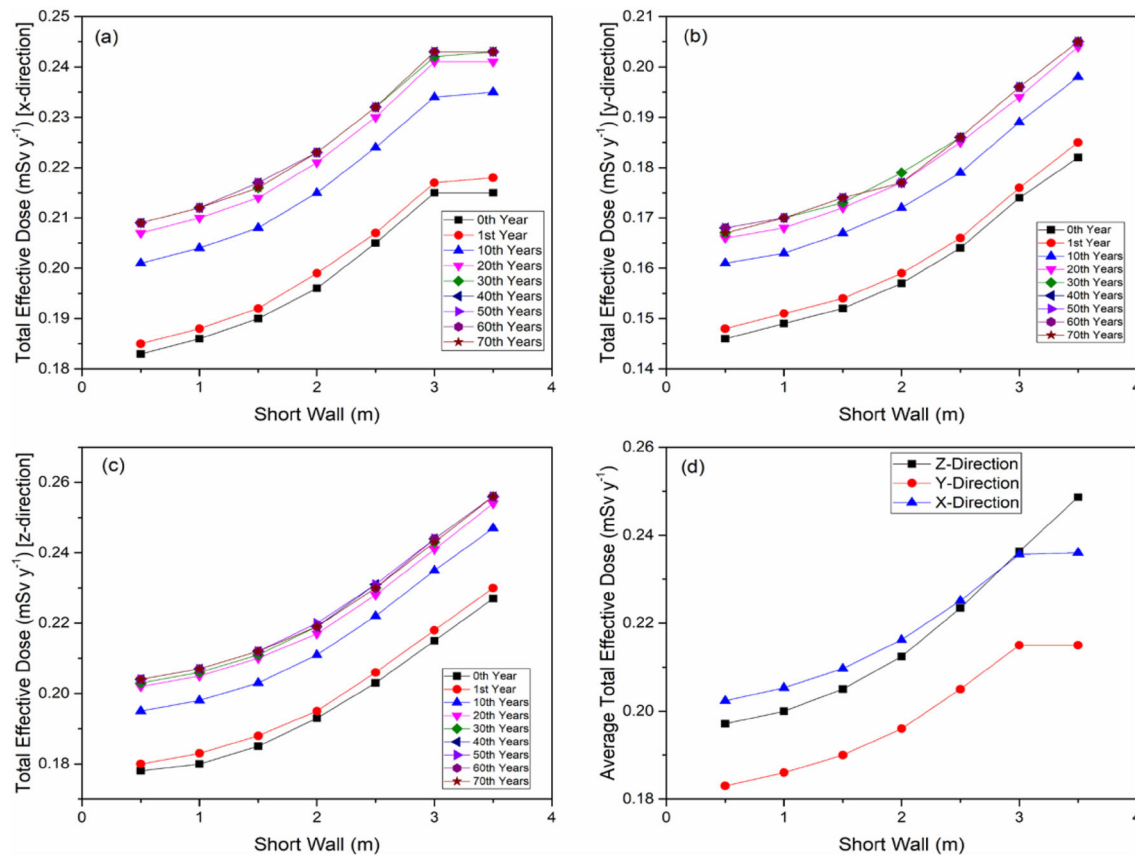
The long-term variations between the TED and ventilation rates are presented in Fig. 5a, b. The duration of the study was 0 to 70 y, and the ventilation rate considered varied from 0.1 to 2.0 h<sup>-1</sup>. The adopted room size was 4  $\times$  5  $\times$  2.8 m<sup>3</sup> and the dweller position was 2  $\times$  2.5  $\times$  1, which were both fixed during the study. The average TED ranged from 0.26  $\pm$  0.01 to 0.61  $\pm$  0.01 mSv/y and decreased as the ventilation rate increased. Stranden [29] reported that the ventilation rate would not be constant over time and would mostly be high during the day due to the operation of normal artificial ventilation systems and opening of doors and windows. The findings of this study agreed with those of previous studies, such as Resica et al. [30] and Stranden [29]. Therefore, ventilation has a significant effect on the TED received by the dweller in the indoor environment [27, 28].

**Fig. 5** Total effective dose and ventilation rate

### 3.3.3 Effect of Position on the TED Received by the Dweller

The effect of position on the TED received by the dweller is presented in Fig. 6a–d. The TED was estimated from various positions and directions ( $x$ ,  $y$ , and  $z$ ) in the room to determine the doses received by the dweller at various positions and directions from the reference wall (in this case, a “short wall”). The TED behaved in an analogous manner from all directions, but the quantity of the





**Fig. 6** (Color online) Effect of dweller position on the total effective dose

TED received by the dweller varies from each direction. It was assumed that the dweller began moving from one end of the room, where the ventilation systems are located, to the other. The TED increased from all directions when the dweller moved away from the ventilation systems until they approached the other end of the room, at which point the doses from the  $x$ - and  $y$ -directions began to behave in a stable manner. The average TED received by the dweller from the  $x$ -,  $y$ -, and  $z$ -directions ranged from  $0.2 \pm 0.01$  to  $0.24 \pm 0.01$ ,  $0.18 \pm 0.01$  to  $0.22 \pm 0.01$ , and  $0.2 \pm 0.01$  to  $0.25 \pm 0.01$  mSv/y, respectively. The percentage variations were 34, 31, and 35%, respectively, and the overall TED received by the dweller was 0.63 mSv/y.

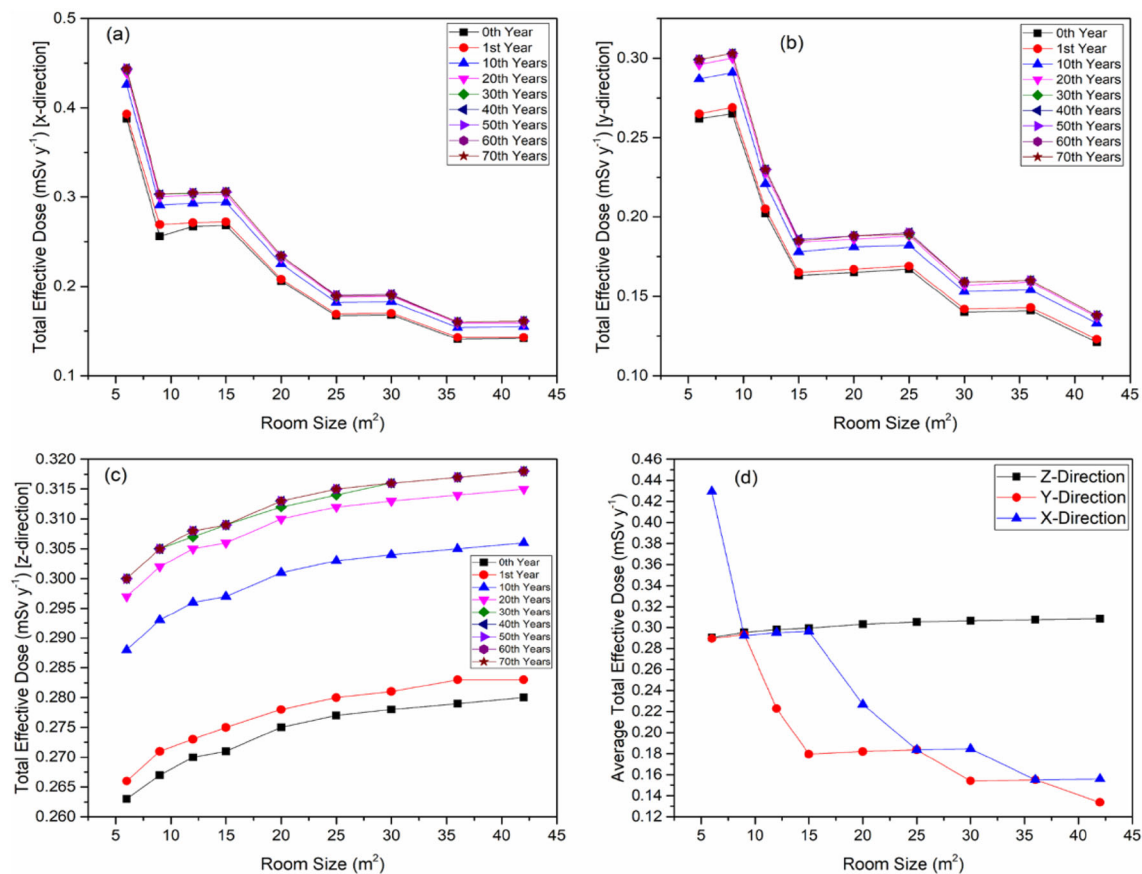
### 3.3.4 Effect of Room Size and Direction on the Total Effective Dose

The long-term radiological dose received by the dweller was assessed using the RESRAD-BUILD computer code presented in Fig. 7a–c, and its variation from 0 to 70 y was investigated. Figure 7d presents the mean variation in the TED received by the dweller with room size, and the

direction of radiation dose in a room significantly influenced the level of exposure received by the dweller.

The variation in the TED from the two walls of the room was approximately 10% and 6% from the  $x$ - and  $y$ -directions, respectively. However, when the contribution of the indoor exposures is considered from the floor and ceiling, the variation in the TED is relatively constant and is approximately 1% from the  $z$ -direction. The simulation results of the TED received by the dweller in a room ranged from  $0.15 \pm 0.01$  to  $0.43 \pm 0.02$ ,  $0.13 \pm 0.01$  to  $0.29 \pm 0.01$ , and  $0.29 \pm 0.01$  to  $0.38 \pm 0.01$  mSv/y for the  $x$ -,  $y$ -, and  $z$ -directions, respectively. The TED received by the dweller from the aforementioned directions is 0.75 mSv/y, and the percentage contribution of the  $x$ -,  $y$ -, and  $z$ -directions is 33, 27, and 40%, respectively. Therefore, estimating TED from one direction in a room would underestimate the TED received by the dweller.

Room size has been proven to be one of the three basic principles of radiation protection, i.e., time, distance, and shielding. It was observed that, when the room size increases, the TED received by the dweller decreases. This suggested that most of the TED was contributed by the walls of the room, which was the reason behind the decreases in the TED values with increasing room size.



**Fig. 7** (Color online) Effect of room size and direction on the total effective dose

However, this is only true when the position of the dweller remains fixed while the room size changes.

The results reported here from the *z*-direction agree with those of previous studies, such as Resica et al. [30] and Majid et al. [31]. However, the variation reported here was only 1%, while that reported by Resica et al. [30] was 6% [30]. Majid et al. [31] estimated the indoor effective dose from a single direction and, therefore, underestimated the TED received by the dweller [31].

#### 4 Conclusion

The activity concentrations of the three essential radionuclides,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , from common ceramic tile materials in Malaysia were studied and analyzed through gamma-ray spectroscopy using HPGe detector. The activity concentrations of the aforementioned radionuclides were assessed for their potential radiological hazards to human health. The activity concentrations ranged from  $38 \pm 0.3$  to  $216 \pm 6$ ,  $42 \pm 0.1$  to  $182 \pm 4$ , and  $350 \pm 30$  to  $1589 \pm 20$  Bq/kg for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , respectively, while the mean values were  $93 \pm 40$ ,  $68 \pm 30$ , and  $673 \pm 200$  Bq/kg, respectively. The mean

activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were all above the typical global values of 50, 50, and 500 Bq/kg, respectively. The calculated  $R_{\text{eq}}$  and  $ACI$  ranged from  $146 \pm 1$  to  $565 \pm 10$  Bq/kg with a mean value of  $241 \pm 83$  Bq/kg and  $0.53 \pm 0.01$  to  $2.04 \pm 0.05$  with a mean value of  $0.87 \pm 0.3$ , respectively. Therefore, the mean  $R_{\text{eq}}$  and  $ACI$  are below the recommended maximum limits of 370 Bq/kg and 6, respectively.

The results of the RESRAD-BUILD computer code simulation for TED to determine the effect of ventilation rate ranged from  $0.26 \pm 0.01$  to  $0.61 \pm 0.01$  mSv/y. The percentage variations in the TED due to dweller position and room size from were 34, 31, and 35% and 33, 27, and 40% for the *x*-, *y*-, and *z*-directions, respectively. The overall TED value received by the dweller due to variations in room size and direction was 0.75 mSv/y. Although the TED value from all directions is still below the action limit of 1 mSv/y, the contribution is significantly different between each direction. The calculated radiological risk parameters were all below the recommended maximum limit. Therefore, the radiological impact of the studied tile materials is negligible.

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