Estimation of the indoor radon and the annual effective dose from granite samples

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Abstract. Inhalation of radon and thoron daughters increases the risk of lung cancer. The main sources of indoor radon are building materials. The aim of this research is to estimate the indoor radon and the annual effective dose from the building materials. Eighteen granite samples bought from the markets in Thailand were measured using an ionization chamber (ATMOS 12 DPX) for the radon concentration in air. Radon exhalation rates were calculated from the radon concentration in chamber. The indoor radon from the granite samples ranged from 10.04 to 55.32 Bq•m⁻²•h⁻¹ with an average value of 20.30 Bq•m⁻²•h⁻¹ and the annual effective dose ranged from 0.25 to 1.39 mSv•y⁻¹ with an average value of 0.48 mSv•y⁻¹. The results showed that the annual effective doses of three granite samples were higher than the annual exposure limit for the general public (1 mSv•y⁻¹) recommended by the International Commission on Radiological Protection (ICRP). In addition, the relationship between the colours and radon exhalation rates of granite samples was also explained.

1. Introduction
Building materials are enriched with radioactive elements and thus are considered as NORMs (naturally occurring radioactive materials). Radioactive elements in NORMs, especially radon (Rn-222) may have an effect on human health. Radon emits high energy alpha particles (5.5 MeV) which can cause damage to the DNA (deoxyribonucleic acid) of human. Therefore, long-term exposure to high levels of radon concentration increased the risk of lung cancer [1-2]. Radon is a radioactive gas and can be released from the building materials into the air by leaking through a crack of the room floor and accumulated in the houses [3-4]. Radon exposure can occur from all building materials made from substances containing significant amount of radium. In some cases, radon levels are very low, but in other instances, building materials may contribute highly to a significant radon exposure. These building materials sometimes contain comparatively high amounts of uranium, radium and thorium, of which radon is a decayed product [5-7]. Reports from many countries as well as Thailand revealed that granite has higher levels of the radon exhalation rate than other building materials [8]. Thus, in this research, eighteen granite samples were bought from several markets in Thailand. Their radon exhalation rates were determined using an ionization chamber (ATMOS 12 DPX) by the closed-chamber method [9]. The rates were calculated based on the measured radon concentration in the chamber. After that, the radon exhalation rates were used for the estimation of indoor radon and the effective dose from the building materials. The resulting indoor radon concentrations were compared with the average of radon background value of 20 Bq•m⁻³ in Thailand surveyed by Moontep et al and Boonyaprapa et al [10-11] and the guidelines

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level for indoor radon of 148 Bq m$^{-3}$ recommended by the United States Environmental Protection Agency (USEPA) [12]. The resulting annual effective doses were also compared with the corresponding results of different sources and the dose limit for the general public of 1 mSv•y$^{-1}$ recommended by the International Commission on Radiological Protection (ICRP) [13]. In addition, we have explained the relationship between the colour index of each granite sample and its radon exhalation rate.

2. Materials and methods

Granite samples (figure 1a) were bought from a building material store in Thailand. These building materials may be imported from aboard or locally produced. Radon in the pore space of any materials will migrate to the atmosphere in a process called exhalation. In this research, building materials with known surface area were estimated for their radon exhalation rate (production rate) using an ionization chamber (ATMOS 12 DPX) by the closed-chamber method. Each sample was put in a closed exhalation chamber made of stainless steel with a volume of 0.05 m$^3$ (figure 2b-c). The closed chamber was tested for the leakage rate by evacuating the container to zero on the vacuum gauge and left for 6 h. Any drop of the vacuum-gauge needle would indicate air leakage. The radon-free air was flowed through an activated charcoal canister into the closed chamber for about 2 min (or until it reached the normal pressure). The closed chamber was kept until the radioactive equilibrium was reached (which normally took more than 30 days).

![Figure 1](image1.png)

**Figure 1.** (a) Colour of the granite samples used; (b) the exhalation chamber; (c) the experimental set up used in this study.

Radon concentration in the closed exhalation chamber was measured by an ionization chamber (ATMOS 12 DPX) (a continuous radon monitor) [14]. An extrapolation of the radon concentration to an infinite time point could be calculated by equation 1. The calculation of radon exhalation rate from building materials using radon concentrations measured in the closed chamber under the radioactive equilibrium state was described by Kovler et al [14] as shown in equation 2:

$$C_t = C_{\infty} \left(1 - e^{-\lambda_{Rn}t}\right)$$  \hspace{1cm} (1)

$$E = \frac{C_{\infty} \lambda_{Rn} V}{S}$$  \hspace{1cm} (2)

where $C_t$ was the concentration of radon measured at time $t$ (Bq m$^{-3}$); $C_{\infty}$ was the maximum radon concentration in equilibrium (Bq m$^{-3}$); $t$ was the accumulation time of the sample (h); $\lambda_{Rn}$ was the decay constant of radon (h$^{-1}$); $E$ was the radon exhalation rate (Bq m$^{-2}$•h$^{-1}$); $V$ was the volume of the chamber (m$^3$), and $S$ was the surface area of the source (sample) (m$^2$).
Radon exhalation rates of the building materials showed a good correlation with indoor radon concentrations under the determining parameters such as the surface area of total wall, the room volume, the outdoor radon concentration and the ventilation rate in the house etc. The estimation of indoor radon concentration in a room due to radon exhalation rates from building materials could be calculated by equation 3 [15-16]:

$$ C = \frac{E S}{V (\lambda_{Rn} + \lambda_v)} + \frac{C_o \lambda_v}{\lambda_{Rn} + \lambda_v} $$

where $C$ is radon concentration (in a room) (Bq•m$^{-3}$); $E$ is radon exhalation rate (Bq•m$^{-2}$•h$^{-1}$); $C_o$ is outdoor radon concentration which is about 5 - 10 (Bq•m$^{-3}$); $V$ is the room volume (m$^3$); $S$ is the surface area of total wall (m$^2$); $\lambda_{Rn}$ is the radon decay constant (h$^{-1}$), and $\lambda_v$ is the ventilation rate at 0.5 (h$^{-1}$).

The room volume and the surface area of total wall in this research were 56 m$^3$ and 90.4 m$^2$ respectively, by referring to the radiation protection report RP-112 [17]. In this research, for the highest safety, the ventilation rate was set at 0.5 h$^{-1}$ [18]. Generally, ventilation is the key factor that affects the indoor radon concentration, and on the average, an air-conditioned room has a higher radon concentration than a non-air-conditioned room in the same category of building. At the same time, the use of the building materials only slightly influences the indoor radon concentration [19].

The indoor radon concentration values estimated were used for the calculation of the annual effective dose due to living in a dwelling. Estimation of the annual effective dose was calculated using equation 4 [20]:

$$ H = C \times F \times T \times D $$

where $H$ is the annual effective dose rate (mSv•y$^{-1}$); $C$ is the radon concentration (Bq•m$^{-3}$); $F$ is the radon-equilibrium factor (0.4); $T$ is the indoor occupancy factor (7000 h•y$^{-1}$); $D$ is the dose conversion factor ($9.0 \times 10^{-6}$ mSv•h$^{-1}$ per Bq•m$^{-3}$).

3. Results and discussion
Radon concentrations from granite samples in a closed chamber under a radioactive equilibrium state, radon exhalation rates, indoor radon concentrations and estimates of the annual effective dose rate are shown in table 1.

The radon concentrations of the granite samples in closed chamber ranged from 14.36 ± 3.31 to 3,400.61 ± 4.39 Bq•m$^{-3}$. The radon exhalation rates ranged from 0.06 to 14.30 Bq•m$^{-2}$•h$^{-1}$. The estimated indoor radon concentrations ranged from 10.04 to 55.32 Bq•m$^{-3}$. The estimated annual effective doses varied from 0.25 to 1.39 mSv•y$^{-1}$. From table 2, the highest radon exhalation rate was found in both pink and orange granite samples, such as GS13, GS 16 and GS18, while the lowest radon exhalation rate was found in both black and white granite samples, such as GS1, GS3, GS4, GS7, GS12 and GS15.

Estimated indoor radon concentration and the annual effective dose rate, in comparisons to the background, the guideline level and the dose limit, were shown in figure 2.
Table 1. The origins of the samples, their colour index, the equilibrium radon concentrations, radon exhalation rates, estimated indoor radon concentrations and the annual effective dose rates.

| No. | Origin of sample | Colour index | Radon concentration (Bq m\(^{-3}\)) | Radon exhalation rate (Bq m\(^{-2}\) h\(^{-1}\)) | Estimated indoor radon concentration (Bq m\(^{-3}\)) | Estimated annual effective dose rate (mSv y\(^{-1}\)) |
|-----|------------------|--------------|-------------------------------------|---------------------------------|-----------------------------------------------|-------------------------------------------|
| GS1 | Thailand         | Black \(^a\), White | 14.36±3.31                           | 0.06±0.01                       | 10.04                                         | 0.25                                      |
| GS2 | Thailand         | Grey \(^a\), Black, Pink | 1061.38±8.84                          | 4.46±0.04                       | 24.04                                         | 0.61                                      |
| GS3 | Thailand         | Black \(^a\), White | 28.65±4.52                            | 0.12±0.02                       | 10.23                                         | 0.26                                      |
| GS4 | Thailand         | Black \(^a\), White | 263.81±9.77                           | 0.80±0.03                       | 12.39                                         | 0.31                                      |
| GS5 | Thailand         | White \(^a\), Grey, Black | 479.33±16.57                          | 2.02±0.02                       | 16.26                                         | 0.41                                      |
| GS6 | Thailand         | Grey \(^a\), White, Black | 744.40±29.08                          | 3.13±0.04                       | 19.80                                         | 0.50                                      |
| GS7 | Thailand         | Black \(^a\), White | 52.33±5.81                            | 0.22±0.02                       | 10.55                                         | 0.27                                      |
| GS8 | Brazil           | Red \(^a\), Black | 1343.01±11.04                         | 5.65±0.05                       | 27.81                                         | 0.70                                      |
| GS9 | India            | Black \(^a\), Grey | 80.97±6.61                            | 0.34±0.06                       | 10.93                                         | 0.28                                      |
| GS10| China            | Black \(^a\), Grey | 60.07±7.84                            | 0.25±0.03                       | 10.65                                         | 0.27                                      |
| GS11| Brazil           | Golden \(^a\), Red, Black | 67.08±6.71                           | 0.25±0.03                       | 10.66                                         | 0.27                                      |
| GS12| China            | Black \(^a\) | 43.60±16.77                           | 0.18±0.07                       | 10.43                                         | 0.26                                      |
| GS13| Vietnam          | Orange \(^a\), Grey, Pink | 3400.61±43.99                         | 14.30±0.02                      | 55.32                                         | 1.39                                      |
| GS14| Finland          | Orange \(^a\), Pink, Black | 521.30±7.73                         | 2.19±0.03                       | 16.82                                         | 0.42                                      |
| GS15| South Africa     | Black \(^a\) | 109.70±9.14                            | 0.46±0.04                       | 11.32                                         | 0.29                                      |
| GS16| Brazil           | Pink \(^a\), Golden, Black | 2672.52±3.88                         | 11.24±0.02                      | 45.59                                         | 1.15                                      |
| GS17| India            | Red \(^a\), Black | 980.63±23.26                          | 4.12±0.10                       | 22.96                                         | 0.58                                      |
| GS18| Brazil           | Pink \(^a\), Golden, Black | 2229.00±41.18                        | 9.37±0.065                      | 39.65                                         | 1.00                                      |

\(^a\) The first colour was the colour index.

Figure 2. Comparison of the estimations of indoor radon concentration (a) and the annual effective dose rate (b) in comparison to the background level of radon, the guideline level of Thailand and the dose limit. The red bars represented black and white granite samples; the orange bars represented pink and orange garnite samples; the black bars represented granite samples of other colours.
Figure 2 showed that the estimations of indoor radon concentration of three granite samples (GS13, GS16 and GS18) were higher than the dose limit for the public. Interestingly, these three granite samples appeared pink and orange in colour. While black and white granite samples produced the lowest radon exhalation rate. Different colours of granites appeared to exhale different amounts of radon. However, additional experiments should be conducted as radon exhalation rates also depend on other factors. Radon exhalation rates also depend on the composition of each granite sample, which contains naturally-occurring uranium and radium. These radioactive elements can lead to a rather high radon exhalation rate. However, from this experiment, differences in the colour of granite samples may be used to assess the amount of the radon exhalation rate from granite. This finding may be useful in the selection of building materials for home owners.

4. Conclusion
In this research, granite samples of multiple origins were bought from a material store in Thailand. The radon concentrations of the granite samples in closed chamber under radioactive equilibrium were monitored by using an ionization chamber (ATMOS 12 DPX). The values of radon concentrations were used for the calculation the radon exhalation rate from granite samples. Estimations of the indoor radon and annual effective dose from the building materials were calculated. The estimations of indoor radon concentration were compared with the background level of Thailand (20 Bq•m⁻³) surveyed by Moontep et al and Boonyaprapa et al [10-11] and the guideline of indoor radon concentration (148 Bq•m⁻³) recommended by USEPA. Estimations of the annual effective dose were compared with the annual effective dose limit for the public (1 mSv•y⁻¹) recommend by ICRP. Radon concentrations from granite samples ranged from 14.36 ± 3.3 to 3,400.61 ± 4.39 Bq•m⁻³. Radon exhalation rates from granite samples ranged from 0.06 to14.30 Bq•m⁻²•h⁻¹. The estimations of the indoor radon concentration ranged from 10.04 to 55.32 Bq•m⁻³. The estimations of indoor radon concentration of six granite samples were higher than the average radon background level of Thailand. However, they were below the guideline level for indoor radon concentrations. Estimations of annual effective dose ranged from 0.25 to 1.39 mSv•y⁻¹. The annual effective dose of three granite samples were higher than the recommended annual effective dose limit for the public. Granite samples with different colours exhaled different amounts of radon. However, additional experiments should be carried out to investigate the relationship between granite colours and radon exhalation rates.

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References
[1] National Council on Radiation Protection and Measurements 1984 Evaluation of Occupational and Environmental Exposures to Radon and Radon Decay products in the United States NCRP Report No. 78 (Bethesda Maryland, MD: National Council on Radiation and Measurements)
[2] World Health Organization 2009 WHO Handbook on Indoor Radon, A Public Health Perspective (Geneva: WHO Press)
[3] Nowak Mo Song B H 1990 Int. Symp. on Radon and Radon Reduction Technology Vol. V: VIII (Atlanta, GA: EPA)
[4] Cothern C R and Smith J E 1987 Environ. Sci. Res. 35 218
[5] Allen J G, Minegishi T, Myatt T A, Stewart J H, McCarthy J F and Macintosh D L 2010 J. Expo. Sci. Environ. Epidemiol. 20 263
[6] Righi S and Bruzzi L 2006 J. Environ. Radioactivity 88 158
[7] Ai-Jarallah M 2001 J. Environ. Radioactivity 53 91
[8] Sola P, Srinurarakul W, Laoharojanphand S and Suwankot N 2014 DAe-BRNS 5th Symp. On Nuclear Analytical Chemistry (NAC-V) (Mumbai: BARC) pp 82-3
[9] Kovler K 2006 Proc. Int. Radon Symp. (Kansas City, MO) pp 31-7
[10] Moontep C, Suparom K, Sola B, Chantarachote W, Wanabongse P and Bovornkitti S 2008 Thamasat Medical J 8 455
[11] Boonyaprapa S, Wanabongse P, Cheepsattayakorn A, Saeung S, Sola B and Bovornkitti S 2008 J. Health Systems Res. 2 460
[12] US EPA 1992 National emission standards for hazardous air pollutants; Emission standards for radon emissions from phosphogypsum stacks; Final rule (40CFR61) Federal Register 57 23305
[13] ICRP 1991 Annals of the ICRP; 1990 Recommendation of the Int. Commission on Radiation Protection (ICRP Publication 60) (Oxford: Pergamon Press)
[14] Kovler K, Perevalov A, Steiner V and Metzger L A 2005 J. Environ. Radioactivity 82 321
[15] Krisiuk E M 1980 Health Phys. 38 199
[16] Quindos L S, Newton G J and Wilkening M H 1987 Radiat. Prot. Dosim. 19 125
[17] European Commission 1999 Radiological protection principles concerning the natural radioactivity of building materials Radiation Protection Report RP-112 (Luxembourg: European Commission)
[18] Krisiuk E M 1980 Health Phys. 38 199
[19] Leung J K, Tso M Y W and Ho C W 1999 Health Phys. 76 537
[20] United Nations Scientific Committee on the Effects of Atomic Radiation 2000 Sources and effects of ionizing radiation 2000 Report to the General Assembly with Annex B: Exposure from Natural Source of Radiation (New York: UNSCEAR)