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Radon Mass Exhalation Rates of Selected Building Materials in Tanzania

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Abstract
This study aimed at determining the mass radon exhalation rate of Tanzania Portland cements and their raw materials for assessment of the radiological hazards due to use of those materials in residential construction. The radon mass exhalation rate was measured by closed chamber coupled with the Pylon AB5 ™ and varied from 0.3 to 13 %. The estimated indoor radon concentrations and annual effective dose for tightly closed standard room were within the safe limits of radon potential health hazards of 600 Bq m^{-3} for dwellings and 1500 Bq m^{-3} for workplaces recommended by International Commission on Radiological Protection (ICRP).
Key words. Building materials; Radon mass exhalation rate; Annual effective dose; Radiological hazard; Tanzania

1. Introduction

Radon ^{222}Rn, thoron ^{220}Rn and actinon ^{219}Rn are naturally existing radioactive noble gases with half-lives of 3.83 d, 55.6 s and 3.96 s respectively. The source of the radioactive gases in the dwellings is the occurrence of the radium isotopes from the uranium and thorium series in the soil, rocks and building materials. From the radiological point of view and due to the half-life period the hazard of the ^{222}Rn is significantly larger than those of ^{220}Rn and ^{219}Rn, therefore ^{220}Rn and ^{219}Rn are omitted in this study. At homes and building offices ^{222}Rn emigrates from the walls, floor and ceilings and can accumulate to harmful levels. The primary concern of the health effects is related to the potential alpha energy concentration and that is exactly associated with the alpha particles emitted from the short–lived decay products of radon such as ^{218}Po and ^{214}Po (UNSCEAR 1988). The atoms of these isotopes at the creating moment often appear in the ion forms chemically reactive and rapidly attach to the aerosols such as particles of dust and water vapor get inside the body through the process of inhalation. When inhaled may be deposited on respiratory tract tissues, decay and emit the alpha particles acting and damaging cells near the deposition site, contributing to increase the risk of lung cancer (Nazaroff 1992; Darby *et al.* 2005).The concentration of the radon daughters in the dwelling air are governed by the radon concentration in the air, but the last is upon to radium concentration in the building materials including with floor, their construction, radon exhalation rate, attachment and recombination, deposition, and transport by diffusion and air ventilation condition and so on (Durrani 1997;Akerblom 1994;Jonsson 1995;Bossew 2003).

The release of the radioactive gas from the materials into the atmosphere is controlled by three processes: emanation, transport and exhalation. The illustration of the mentioned processes is presented in Figure 1. Emanation is the escape of the radon gas from the solid medium into the air filled pore spaces (interstitial space), governed by the momentum and recoil energy of alpha particles from the radioactive decay of radium. The recoil distance for ^{222}Rn ranges 20-70 nm in common minerals, 100 nm in water, and 63µm in air (Nazaroff 1992). From the pores spaces (interstitial space), radon can migrate through diffusion process or seepage. The diffusion of radon is a process controlled by radon gas concentration gradient across the radon gas sources (rocks, soils, building materials, and other different materials) and the surrounding air. Finally, a fraction enters the atmosphere before undergoing radioactive decay the process known as exhalation. The exhalation rate is the radon fraction in the free air originated from the radon created by the radium decays in the inside of the materials. It was revealed that, the value of exhalation rate depends on air condition and on the physical properties of the material samples such as moisture, grain sizes, and so on (De Martino 1998). The radon exhalation rate ranges from 0 (in the case of no radon release from the material), to 1 (where all radon escapes).

A number of studies have been conducted to establish the radium concentrations in raw and industrial building materials and their radon exhalation rate (EC 1999; Hassan *et al.* 2009; Sakoda *et al.* 2010; IAEA 2013). The ^{226}Ra concentration of the most typical building materials in the World is summarized in Table 1 (IAEA 2002).

The Portland cements are typically the products derived from sedimentary or weathered rocks such as: sandstones, clay, limestone, shale, gypsum, pozzolan (UNSCEAR 2008). Portland cement clinker is manufactured by heating a calcareous material, typically limestone, and an argillaceous like clay or shale. The final product of Portland cement is gained by intergrading gypsum (sulphate minerals) with the clinker (Taylor 1997). The production processes also influences the radionuclide activity concentration of final products and thus determine the activity concentrations in the construction materials. Generally, the radium concentration in the final product should be controlled by the radium activity of the raw materials and calculated by formula:

(1)

Where $Ra_{prod.} = \sum_{i=1}^{n} w_i Ra_i$ $Ra_{prod.}$ and Ra_i is the concentrations of radium, in cement and i-th raw material composition respectively; w_i – weigh contribution of the i-th composition.

The aim of this work is to determine the ^{222}Rn mass exhalation rates of Tanzania cements and selected building materials by closed chamber method and to estimate their contributions to indoor ^{222}Rn concentration. The obtained results were used for assessment of the adequate radiological indices, and appraising the investigated building materials from the radiological point of view.

2. Materials and Methods.

A number of methods have been developed to measure the radon exhalation rates of building materials including the accumulation method, the charcoal method, the SSNTD (solid state nuclear track detector method (Limoto *et al.*2008) and the closed chamber method (CCM). All methods have one purpose in common that is measuring radon concentration in a closed space after the radioactive secular equilibrium between radon and radium is achieved. In this study the closed chamber method was adopted using the radon chamber coupled with radon meter "Pylon AB5™ (Figure 2).

The four brands of cement available in Tanzania trademarked as Tembo, Simba, Rhino and Kilwa, collected from the local market were examined in this study. The other selected building material included pozzolan; occasionally used for plastering (with few centimetres thin wall) in local communities, gypsum as thin (1-3 cm) gypsum boards for walls and ceilings and clay and sandstone for fired and unfired bricks. Pozzolan, clay, sandstone and gypsum were collected from their respective quarries in Songwe and Kilwa located in Mbeya and Lindi regions respectively. The samples were collected according to IAEA, (1989) standard sampling procedures for environmental sampling; the localizations of the collected samples are shown on Figure 3.

The sample under investigation weighed from 0.5 to 1 kg were milled until the grain sizes lower than 1 mm and dried at 110°C for 24 hours and placed inside the exhalation chamber of volume of 10 dm^3 and tightly sealed to prevent escape of radon. Each enclosed sample was left for one month to establish the equilibrium between the ^{226}Ra and ^{222}Rn. After attainment of the equilibrium the closed chamber was connected with AB-5 Pylon coupled with the Lucas cell by using the plastic pipes and valves (see Figure 2). The inlet and outlet located on the top of the chamber with the inlet tube extended down to the bottom of the chamber.

The AB-5 is built in pump that circulates the air through circuit connecting the chamber and Lucas cell. For establish the uniform distribution of the air in the circuit and to gain the minimum uncertainty, the AB-5 was allowed to pump the air for 30 minutes and counts were collected for several runs, every counting run was lasting for 2 hours. The total activity of radon released from the sample material into the air is evaluated by measured radon content in the Lucas cell and the effective volume of the measurement system. The effective volume is determined by considering the volumes of the chamber, closed vessel, all pips inside the closed vessel, the loop system for radon measurement and of the Lucas cell (IAEA 2013). The total Rn produced in the closed chamber was obtained by multiplying the sample mass and adequate radium activity of the investigated material. The mass exhalation rate therefore is given by the following expression (IAEA 2013).

$$E_m = \frac{C_d}{mRa} \quad (2)$$

Where E_m is the mass exhalationrate;
m is the mass of the sample (kg);
and Ra is the radium activity concentration (Bq kg^{-1}) of the investigated material.
C_d is the radon content in the sampling device (Bq) and calculated using formula $C_d = (C_L \cdot V_d)/V_L$
where V_d, V_L are the effective volume of the sampling device and Lucas cell respectively;
C_L – radon content in Lucas cell, and calculated by formula: $C_L = NCR/(3 \cdot \varepsilon \cdot t)$, where ε - recorded alpha efficiency of AB-5 device and t – measurement time in seconds;
NCR – net count rate obtained by subtraction the measured count rate after pumping from the background one (the count rate measured for the same Lucas cell before pumping air). The background measurements were performed before pumping for the used Lucas cell. The background measurement time was predicted for the relative uncertainty of count rate lower than 2 %.

The mass exhalation rate was then used to determine the potential radon concentration originating from the building materials in the air inside the "standard room" of 4×5×2.8 m (56 m^3) with door of 1 m × 1.8 m and window of 1.4 m × 1.4 m dimensions according to the EC Standards (EC1999). The wall thickness is often upon on the application of the building materials. For instance the thickness of the wall built of gypsum is assumed to be 3 cm, of pozzolan and soil 1 cm each, cements bricks 20 cm and that from clay and sandstone bricks the thickness amounts to10 cm each (EC1999). The radon contribution from the building material was estimated as follows

$$C_{Rn} = \frac{A_{Ra} \cdot E_m \cdot M}{V} \quad (3)$$

where C_{Rn} is concentration of the radon originating from the building material, A_{Ra} and E_m are the activity concentration of radium and mass exhalation rate of the building material respectively. V is the volume of the room and M is the mass of the walls and estimated by formula (2×2.8× (4+5) – (1x1.8+ 1.4x1.4)) m^2 x a (m) ×ρ (kg m^{-3}) for standard room; where a is the wall thickness and ρ is the building material density. The density of every investigated material was determined by weight and the sample volume of the milled material and summarized in Table (3).

The radiological hazard is directly connected with the potential energy alpha concentration originated from the alpha particles emitted from the decay of radon progeny and expressed in J/m^3 or Working Level (WL) unit (WHO 2009). The worker's/dwellers exposure to radon daughters is expressed in units of Working Level Months (WLM). Working level month is equivalent to the exposure at an average concentration of 1 WL for 170 working hours (WHO 2009; ICRP 1993).

The annual potential alpha energy potential expressed in WLM for dwelling habitant is estimated by formula (Sharma *et al.* 2012):

$$E(WLM/y) = \frac{8760 \times n \times F \times C_{Rn}}{170 \times 3700} \quad (4)$$

where n is the fraction of time spent indoors and arbitral assumed to be equal to 0.8; F is the equilibrium factor between Rn and its progeny and assumed to be equal to 0.42; C_{Rn} – radon concentration in the air [Bq m^{-3}], 8760 is the number of hours per year; and 170 is the number of hours per working month (UNSCEAR 2000). The annual potential alpha energy can be translated to the annual effective dose by multiplication of the alpha energy potential and a conversion factor of 3.88. By this way the annual effective dose is expressed in mSv (ICRP 1993). The uncertainty of radon mass exhalation rate can be calculated using the equation (2) and propagation low as follows.

$$\frac{uE_m}{E_m} = \left[\left(\frac{uC_d}{C_d} \right)^2 + \left(\frac{uM}{M} \right)^2 + \left(\frac{uR_a}{R_a} \right) \right]^{1/2} \quad (5)$$

Where uE_m, uC_d and uRa are the uncertainties of measured radon content in the chamber, sample mass and ^{226}Ra concentration of the investigated material.

3. Results and Discussion

The measured radon mass exhalation rates, their uncertainties, Rn concentrations and annual effective doses estimated for the Standard built from the investigated materials are summarized in Table 2.

The radon mass exhalation ranges from 0.3 to 13 % for raw materials and from 0.7 to 2.8 % for cements. The variation in the mass exhalation rate of cements is due to radioactivity characteristics of the individual cements and mineral structure of their components.

The indoor radon concentrations and annual effective doses estimated for hermetically closed “standard room” with the assumption that the exhaled radon was only released from its walls built from the investigated materials. The calculated indoor radon concentration and annual effective dose ranged from 3.1 to 1012.3 Bq m^{-3} and from 0.05 to 17.5 mSv respectively. These values were within the safe limits of radon potential health hazards of 600 Bq m^{-3} for dwellings and 1500 Bq m^{-3} for workplaces corresponding to annual dose limits of 10 and 20 mSv respectively (ICRP 1993, 2009).

It is customarily understood that the radon content in the air is the quantity of radium in the building materials (Figure 4).

However, some discrepancies were observed due to other parameters such as mineral structure, chemical composition and so on. For instance, the slight elevation of the mass emanation rate and annual effective dose exposure from Tembo cement should be connected with radium quantity and its distribution in the volcanic pozzolan and clay. Due to characteristics of the adsorption processes the distribution of the radium atoms in the pozzolan and clay may be closer to the particle surfaces and this orientation enhances the radon escape from the grain space to the air. It was observed that radon exhalation rate from the soil was significantly lower than the rest of the materials; this fact can be connected with the occurrence of organic fractions in the soil, which often contains a lot of the water particles in their cell structures. Water can make the shield for Rn atoms escape from the solid material to the air (Bossew 2003).

4. Conclusion

Radon mass exhalation rates of Tanzania cements, gypsum and pozollan have been determined by closed chamber method. On the basis of the obtained results, the indoor radon contribution of each individual material has been estimated. All of the calculated indoor radon exposure values are less than the international recommended indoor

^{222}Rn exposure limits. The annual effective doses were also determined and found to be within the permissible range. The difference in radon mass exhalation rates supports the suggestion that radon emanation is a function of Ra concentration, moisture and particle structure of building materials. Based on the measured mass emanation rate, calculated Rn concentration and related effective dose one can states that the investigated materials are safe for the dwellers whenever used for construction purpose.

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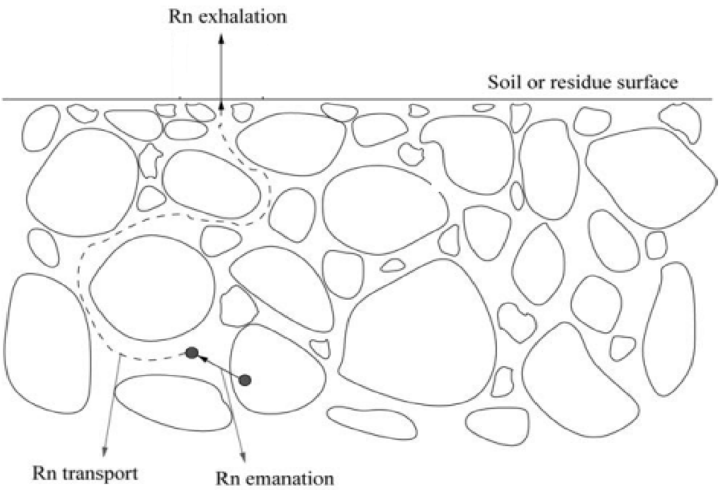

Figure 1. Processes leading to the radon release to the atmosphere (IAEA, 2013)

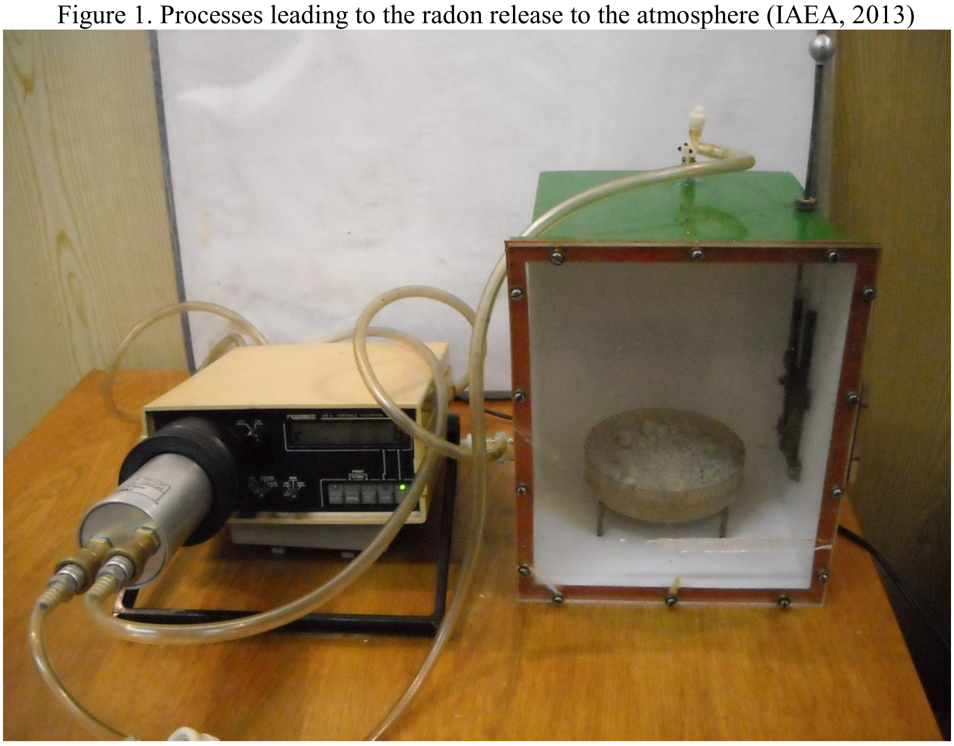

Figure 2. The view of the measurement device

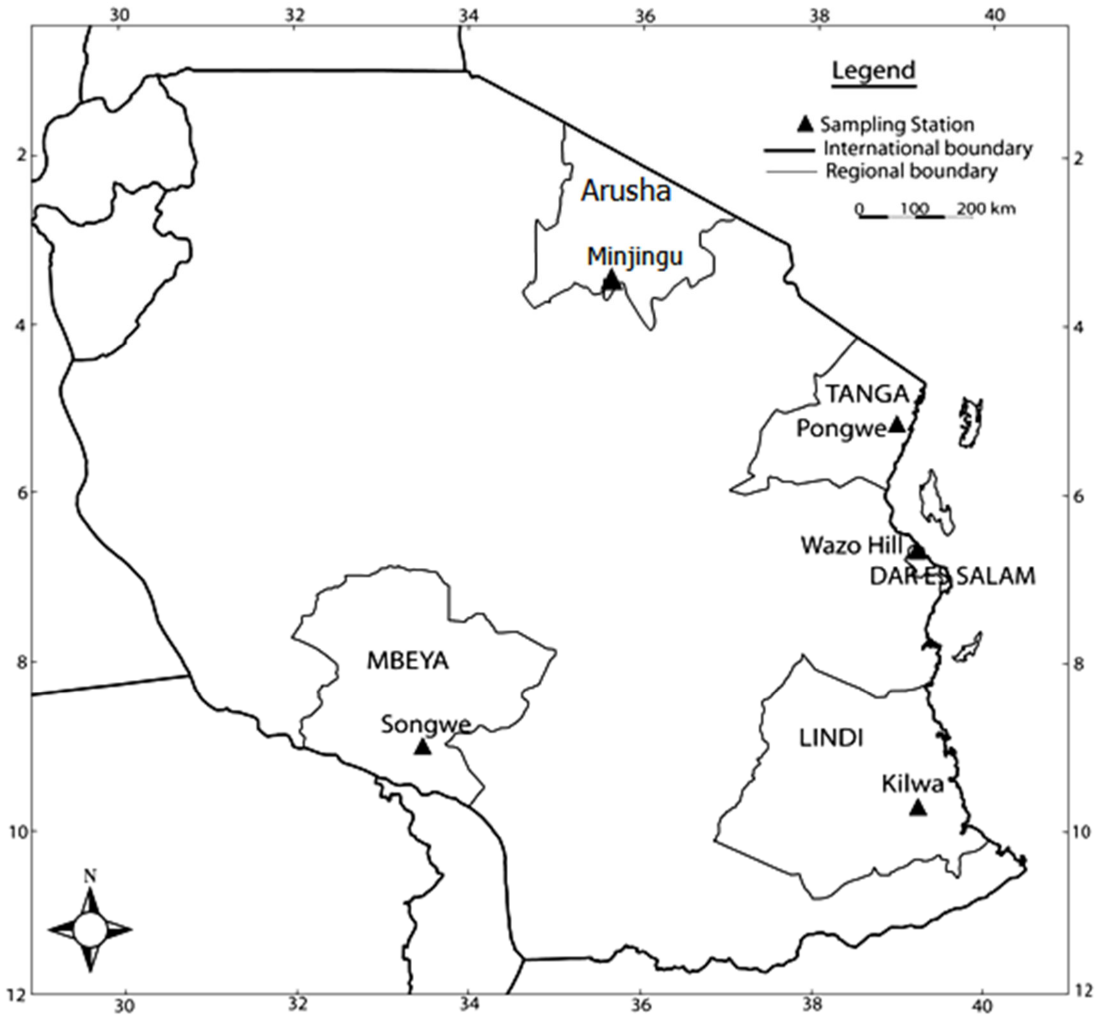

Figure 3. Sketch of Tanzania showing sampling sites (names of the regions in capital letters)

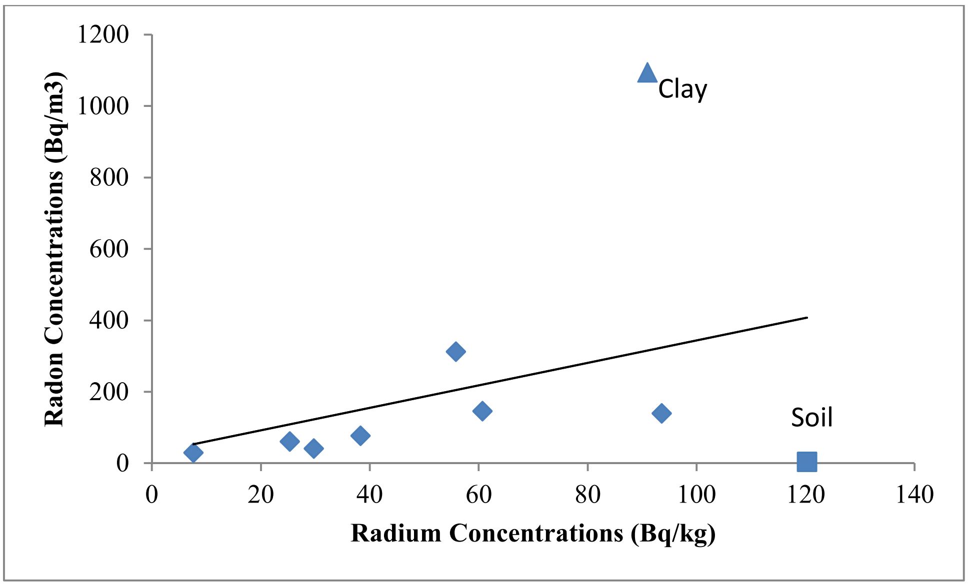

Figure 4. A graph of Rn concentration in the air versus Ra concentrations in the sample material

Table 1 Concentration ranges ^{226}Ra of the most typical building materials in the World (IAEA, 2002)

Material	^{226}Ra (Bq kg^{-1})	Material	^{226}Ra (Bq kg^{-1})
Concrete	1-250	Cement	7 – 180
Clay (red) bricks	9 - 2200	Tiles (glazed and unglazed)	30 – 200
Sand-lime bricks/limestone	6 – 50	Phosphogypsum plasterboard	4 – 700
Natural building stones	1 – 500	Blast furnace slag stone and cement	30 – 120
Natural gypsum	< 1 - 70		

Table 2 The estimated Rn concentration and annual effective doses resulted from the released radon from the wall built from investigated materials

Material	^{226}Ra (Bq kg^{-1})[1]	Density (kg m^{-3})	Net count rate per 2h	E_m (%)	Rn (Bq m^{-3})	Annual effective dose mSv
Rhino cement	60.7±2.7	1112	124±33	1.2±0.3	134.9	2.3
Tembo cement	55.8±4.3	1187	327±52	2.8±0.4	308.9	5.3
Kilwa cement	29.7±4.0	1106	51±30	0.7±0.4	38.3	0.7
Simba cement	38.3±3.9	1123	107±27	1.0±0.3	71.6	1.2
Pozzolan	93.6±1.3	1385	4500±903	12.0±2.4	129.6	2.2
Soil	120.3±1.6	1023	120±26	0.3±0.1	3.1	0.05
Gypsum	7.6±0.4	1105	119±22	13.0±2.4	27.3	0.5
Sandstone	25.3±0.7	1347	193±27	2.0±0.1	56.7	0.9
Clay	91.0±1.3	1113	3750±612	12.0±0.02	1012.3	17.5

[1] the ^{226}Ra concentrations of the investigated materials were measured by gamma spectrometry (Amasi *et al.* 2014)