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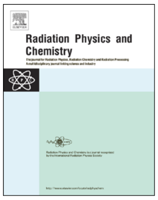

Radon exhalation from building materials used in Libya

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HIGHLIGHTS

- Radon exhalation was measured in building materials (BM) by the can technique.
- The results are mostly within the worldwide range of values found in BM samples.
- Two high values of radon concentration have been observed from granite and marble.
- No significant risk to the human beings due to the presence of radon in the homes.

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ABSTRACT

Radon exhalation rates have been determined for various different samples of domestic and imported building materials available in the Libyan market for home construction and interior decoration. Radon exhalation rates were measured by the sealed-can technique based on CR-39 nuclear track detectors (NTDs). The results show that radon exhalation rates from some imported building materials used as foundations and for decoration are extremely high, and these samples are the main sources of indoor radon emanation. Radium contents and annual effective doses have also been estimated.

1. Introduction

Radon (^{222}Rn) is a part of the natural ^{238}U series decay chain and is the only gas to be found in the series. Ultimately, it decays through a series of short-lived progeny, with short half-lives ranging from μs to minutes, until it reaches ^{210}Pb with a half-life of 22.3 years. These radioactive elements can be found in almost all types of soils, such as rocks, granite, brick, sand, cement, and gypsum, from which building materials are produced, as well as marble and ceramics for decorative use. Radon (^{222}Rn), as an emitter of α-particles with energy 5.48 MeV, is the most crucial and dangerous radioactive gaseous element in the science of environmental radioactivity. This noble gas, although not so noble in its health hazard effect, is chemically inert and can move through the Earth and structural materials with a half-life of 3.82 days to reach the outdoor air and the indoor environment (Nazaroff and Nero, 1988). ^{226}Ra, with a half-life of 1600 years, decays to ^{222}Rn by emitting α-particles and is inherently present in the above-mentioned building materials.

In general, part of the natural redistribution of ^{222}Rn in the environment is a portion exhaled from the soil (Saad et al., 2002, 2013; Saad, 2008) and building materials (Abu-Jarad et al., 1980; Mustonen, 1984; Al-Jarallah, 2001; Al-Jarallah et al., 2001; Saad et al., 2010), which disperses into the atmosphere. The radiation dose from inhaled decay products of ^{222}Rn is the dominant component of natural radiation exposures of the general population. However, ^{222}Rn is reported as a contributor of the largest component of human exposure to natural radiation (UNSCEAR, 2000). Recently, several interesting studies of people exposed to radon have confirmed that radon in homes and workplaces represent a serious health hazard (European Commission (EC), 1999; WHO, 2005, 2009). The exposure of people to high concentrations of indoor radon for long periods causes pathological effects and functional respiratory changes, which consequently lead to an increased risk of developing lung cancer. This risk depends on the concentration of radon indoors, the duration of exposure, and the degree of ventilation in the houses (Webb, 1992; Lubin and Boice, 1997; Neuberger and Gesell, 2002; Lazar et al., 2003). Although radon is formed mainly in the rock and soil upon which a house is built,

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exhalation from building materials is one more potential source of radon in the indoor environment (Cothern and Smith, 1987; UNSCEAR, 2000, 2008; WHO, 2009; EPA, 2009; Saad et al., 2010; Chen et al., 2010; Bavarnegin et al., 2012).

The measurements of radon exhalation rates from building materials used in Libya have been limited (Saad et al., 2010). Building materials, domestic and/or imported, generally used in Libya have no special problem from the radioactivity point of view, but the main potential problem arises when materials with relatively high radioactivity, especially imported granite, ceramic, and marble materials, are used decoratively indoors. Fortunately, the number of such materials is small, and they can be replaced by alternative materials from other countries. The main goal of the current study has been to determine radon exhalation rates from building materials used in Libya. Annual effective doses could thereby be estimated. The results are compared with those obtained by many scientific research groups in other parts of the world.

2. Theoretical details

The radon level of a construction material sample, placed in an emanation container or mounted on it, can be monitored with CR-39-based NTDs. The radon concentration in the closed can or emanation container reaches secular equilibrium within several half-lives of radon. It should be noted that the radon exhalation rate is constant, whereas the radon concentration reaches a maximum value that depends on the exhalation rate. A radon detector based on CR-39 NTDs can be used to measure the accumulated activity and then to evaluate the average radon exhalation rate from building materials or soil using the following equations (Fleischer and Mogro-campero, 1978; Khan et al., 1992; Mahur et al., 2008a, 2008b; Saad et al., 2010, 2013):

$$E_A = \frac{C_{\mathrm{Rn}}\lambda V}{A[T + 1/\lambda_{\mathrm{Rn}}(e^{-\lambda_{Rn}T} - 1)]} \tag{1}$$

$$E_M = \frac{C_{\mathrm{Rn}}\lambda V}{M[T + 1/\lambda_{\mathrm{Rn}}(e^{-\lambda_{Rn}T} - 1]} \tag{2}$$

where E_M is the mass exhalation rate (mBq kg^{-1} h^{-1}), E_A is the areal exhalation rate (mBq m^{-2} h^{-1}), C_{Rn} is the integrated radon exposure (Bq m^{-3}), A is the total surface area of the building material sample from which radon is exhaled (m^2), V is the empty volume of the emanation container or can (m^3), M is the mass of the building material sample (kg), λ is the decay constant of radon (h^{-1}), and T is the time since sealing (h).

The risk of lung cancer from domestic exposure due to radon and its daughter nuclides can be calculated directly from the effective dose equivalents. The radiation hazards due to radon and its daughter nuclides are estimated from the radon exhalation rates of building material samples. The contribution of indoor radon concentration from building materials and soil can be calculated from the following formula (Nazaroff and Nero, 1988; Mahur et al., 2008a, 2008b; Saad et al., 2010, 2013):

$$C_{\mathrm{Rn}} = \frac{E_x \times S_r}{V_r \times \lambda_V} \tag{3}$$

where C_{Rn} is radon concentration in construction materials contributing to indoor radon, E_x is radon exhalation rate (Bq m^{-2} h^{-1}), V_r is room volume (m^3), and λ_V is air exchange rate (h^{-1}). In these calculations, the maximum radon concentration from building materials was assessed by assuming the room to be a cavity with the ratio $S_r/V_r = 2.0$ m^{-1}, where S_r and V_r are the internal surface area and volume of the room, respectively, and the air exchange rate λ_V was taken to be 0.5 h^{-1}. The annual effective dose equivalent E_p is then related to the average radon concentration C_{Rn} and is given by the following formula:

$$E_P\left[\mathrm{WLM}\ y^{-1}\right] = \frac{8760nFC_{\mathrm{Rn}}}{170 \times 3700} \tag{4}$$

where C_{Rn} is in Bq m^{-3}, n is the fraction of time spent indoors, F is the equilibrium factor, 8760 is the number of hours per year, and 170 is the number of hours per working month. Values of $n = 0.8$ and $F = 0.42$ were used to calculate E_p. The effective dose equivalents from radon exposure were estimated by using a conversion factor of 6.3 mSv/WLM (International Commission on Radiological Protection (ICRP), 1987).

The effective radium content in the building material samples was determined by using a track detection method (Somogyi, 1986). Once radioactive equilibrium between radium and radon is established, one may use the analysis of the radon α-activity for the assessment of the steady-state activity concentration of radium. After sealing of the can, the activity concentration of radon begins to increase with time T as follows:

$$C_{\mathrm{Rn}} = C_{\mathrm{Ra}}(1 - e^{-\lambda_{Rn}T}) \tag{5}$$

where C_{Ra} is the effective radium content of the building material sample. This effective radium content of a specimen in a sealed can be calculated as

$$C_{\mathrm{Ra}}(\mathrm{Bq\ kg^{-1}}) = (\rho/kT_e)(hA/M) \tag{6}$$

where h is the distance between the detector and the top of the solid sample in m, ρ is the counted track density, k is a calibration factor of the CR-39 track detector, and T_e denotes the effective exposure time given by

$$T_e = T - 1/\lambda_{\mathrm{Rn}}(1 - e^{-\lambda_{Rn}T}) \tag{7}$$

The exposure time in these measurements was 90 days.

3. Experimental procedure and set-up

A total of 37 samples of building materials were collected from different companies and stores in Benghazi, comprising 17 tile samples, 15 slab samples, and 5 porous powdery building material samples. The samples of slabs and porous powdery materials were dried in an oven at a temperature 105 ± 1 °C for 24 h to remove moisture. For coding, the slab samples that were thermally treated are designated as dry samples, whereas tile samples not subjected to any thermal drying are designated as non-dried samples. They were obtained from different stores, and were therefore treated as different samples. Each sample, porous powder or slabs with different weights, was placed in a cylindrical stainless steel container of radius 7.35 cm, length 14.8 cm, and volume 2.513×10^{-3} m^3. In addition, this container was also placed inversely on the surface of the tile samples of building materials. The material in the form of a porous powder was gently pressed to form a disk-like shape, which permitted radon to diffuse out of this host material with a high degree of homogeneity. The thickness of each disk-like sample in the emanation container was about 2 cm. The emanation container was tightly sealed to prevent the escape of radon and was kept sealed for 3 months to obtain good statistics, after which it was ready for α-particle measurements using a CR-39 nuclear track detector (NTD). The concentration and exhalation rate of radon can be determined using CR-39 detectors because of their ability to register tracks at different levels of sensitivity. The CR-39 polymer sheets of TASTRAK were produced and provided by Track Analysis Systems Ltd. (TASL), Bristol, UK. The standard thickness of 750 μm was used. The polymeric detector samples for the present study were cut to a size of 1.5 × 1.5 cm^2. Prior to exposure to the radiation from radon, the protective polyethylene cover was removed. The emanation container was sealed with silicone and stored for 90