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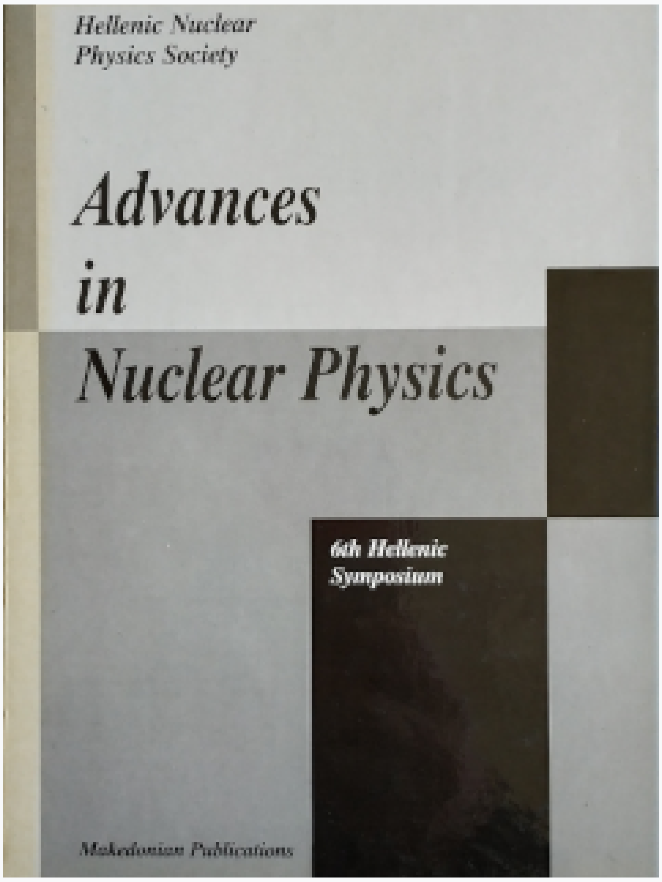

A Research Study on the Radiological State of Building Material and the Risk Assessment

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A Research Study on the Radiological State of Building Material and the Risk Assessment

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Abstract

The objective of the present study is focused on the assessment of the radiological risk from building materials used in Attica region, Greece. Bricks and concrete commonly used in Attica region have been studied for both natural radionuclide content and radon exhalation. The high-resolution gamma-ray spectroscopy technique, as well as radon exhalation measurements, have been employed. The technique used for the measurements of the radon exhalation is called "continuous accumulation-counting" method and has been described in detail by the authors in earlier publications. This measurement technique is based on the continuous air flow through a sealed chamber containing the sample and through a scintillation counter (Lucas cell). The counting is continuous as well.

1 Introduction

The radionuclides occurring naturally in building materials are sources of external and internal radiation exposure in dwellings. The external radiation exposure is caused by the gamma radiation originating from members of the uranium and thorium decay chains and from ^{40}K. The internal radiation exposure, mainly affecting the respiratory tract, is due to the short-lived daughter products of radon which exhales from building materials into room air. Radon ^{222}Rn is a noble gas that belongs to the uranium ^{235}U radioactive series. It is an alpha decay radioactive nucleus and its half life is 3.8 d. The radon exhalation in the environment is the result of ^{226}Ra decay in the elementary grains of a solid body that contains radium. A part of the produced radon due to regression gets into the pores of the body and by diffusion it is possible to reach surface and escape into the environment. The fraction of radon that gets into the pores characterizes the particular body and is called radon emanation coefficient. The radon that gets into the pores does not reach always the surface. The part of radon that decays in its way to the surface depends on the effective diffusion coefficient of radon in the specific medium $D = \varepsilon\lambda l^2$ where ε is the porosity of the medium, λ is the decay constant of ^{222}Rn and l is the radon diffusion length in the medium. The average annual effective equivalent dose from radon inhalation is of the order of 1 mSv which is the 50 % of the doses caused annually by the natural sources (UNSCEAR 1988). The main radon sources are soil and building materials that contain certain amounts of ^{226}Ra. Because of radon accumulation in room air, radon inhalation indoors gives the most significant contribution to the total dose caused by radon.

2 Materials and methods

Brick samples have been collected from major local producers, representing 80 % of the total production in Attica. Concrete was sampled from twenty randomly selected resent buildings in Attica.

2.1 Radon exhalation rate

The technique used for the measurements of the radon exhalation is called "continuous accumulation-counting" (Savidou et al. 1996) This measurement technique is based on the continuous air flow through a sealed chamber containing the sample and through the scintillation counter (Lucas cell) and the counting is continuous as well. The concentration of radon and its daughters in the counter increases with time. The accumulated counts are proportional to the time integral of the total alpha-activity in the counter. To prevent the counting of thoron (^{220}Rn), the samples are covered hermetically with a thin PVC membrane. The ration of samples volume to the volume of the accumulation chamber has been 1:6 and the air flow rate has been 2 $l \cdot min^{-1}$. The low limit (3 σ of the background) of the radon emanation rate for counting time 24 h is 7 $mBq \cdot h^{-1}$.

All the measurements have been carried out in laboratory conditions, in which the temperature is maintained at 20 °C and the relative humidity at 50 %, so significant effects due to changes in ambient conditions have been avoided. The samples have been kept for more than three months under laboratory conditions.

2.2 Specific activities

The concentrations of the natural radionuclide ^{226}Ra, ^{232}Th and ^{40}K are determined by the use of high-resolution gamma spectrometry. The samples are powdered, closed in 75 cm^3 sealed containers and kept about 3 weeks before measurements in order to achieve radioactive equilibrium between ^{226}Ra, radon and its short-lived decay products. The concentration of ^{226}Ra is derived from the 295 keV and 352 keV photopeaks of ^{214}Pb and the 609 keV photopeak of ^{214}Bi as the mean value. The 186 keV photopeak of ^{226}Ra is not used because of the interfering peak of ^{235}U, with the energy of 185.7 keV. The ^{232}Th concentration is determined from the 583 keV and 911 keV photopeaks of ^{208}Tl and ^{228}Ac respectively. The ^{40}K concentration is determined from its 1460 keV photopeak. The counting time is 12000 s.

3 Theoretical estimation of radon concentration indoor

3.1 222Rn exhalation rate from a wall

The differential equation (Culot M. V. J. et. al. 1976) which describes the diffusion in the porous medium is :

$$\frac{D}{\varepsilon \cdot u} \cdot \frac{\partial^2 \Phi(x)}{\partial x^2} - \frac{\lambda}{u} \cdot \Phi(x) + f = 0 \quad (1)$$

where Φ(x): Radon flux per unit area of porous medium (Bq $m^{-2}h^{-1}$), λ: Radon decay constant (h^{-1}), f : Radon production rate into the interstitial space per unit of volume of the interstitial space (Bq m^{-3} h^{-1}), u: Mean velocity of radon atoms in the medium (m h^{-1}), ε: Porosity of the medium.

Radon production rate is defined by the equation (Stranden E. et. al. 1980):

$$f = \frac{\lambda \cdot C_{Ra} \cdot \rho \cdot n}{\varepsilon} \quad (2)$$

where C_{Ra}: Radon concentration per unit mass of the material (Bq kg^{-1}), ρ: The density of the material (kg m-3), n : Radon emanation coefficient of the material.

The mean velocity of radon molecule in the medium (m h^{-1}) is defined by the equation:

$$u = 36 \cdot \sqrt{\frac{8k \cdot T}{\pi \cdot m}} \quad (3)$$

where m : The mass of radon molecule (g), k : The Boltzman constant (1.38×10^{-16} erg ^{0}K), T : The temperature of Radon gas (^{0}K).

In the case that the study concerns the diffusion of radon in a wall, the solution of the differential equation (1) is

$$\Phi(x) = A \cdot e^{x/l} + B \cdot e^{-x/l} + \frac{f \cdot u}{\lambda} \quad (4)$$

By using the boundary conditions mentioned in (Raptis et. al. 1996)

$$\Phi(x) = 2 \cdot A \cdot \cosh\frac{x}{l} + \frac{f \cdot u}{\lambda} \quad (5)$$

where

$$A = \frac{-\frac{f \cdot u^2}{4\lambda}\left[\frac{u}{2}\sinh\frac{d}{2l} + \frac{\varepsilon \cdot \lambda \cdot l^2}{2}\cosh\frac{d}{2l}\right]}{\left[\frac{u^2}{8} + \frac{(\varepsilon \cdot \lambda \cdot l)^2}{2}\right]\sinh\frac{d}{2l} + \frac{\varepsilon \cdot \lambda \cdot l \cdot u}{2}\cosh\frac{d}{l}} \quad (6)$$

The exhalation rate of radon from the surface of a wall is evaluated by

$$F = -S \cdot D \cdot \frac{\partial C(x)}{\partial x}\Big|_{x=\frac{d}{2}} \quad (7)$$

So the exhalation rate of radon from a surface S is given by

$$F = -2 \cdot S \cdot A \cdot \varepsilon \cdot \lambda \cdot l \cdot \sinh\frac{d}{2l} \quad (8)$$

Radon concentration C(t) = Φ/u indoors depends on the following factors: a) Radon exhalation rate per unit area from the wall, the ceiling and the floor of the room (Bq m^{-2} h^{-1}), b) The ventilation rate (h^{-1}) of the room, c) Radon concentration outdoors (Bq m^{-3}), d) The ratio of the total area of the surfaces to the volume of the room (m^{-1}).

Radon concentration in the room is considered uniform. The differential equation that describes radon concentration in room air at equilibrium conditions (Porstendorfer J. et. al. 1978) is:

$$\frac{\sum F}{V} + C_{out} \cdot u - C_o \cdot \lambda - C_o \cdot u = 0 \quad (9)$$

where Fi : Radon exhalation rate from the walls, the ceiling and the floor (Bq m^{-2} h^{-1}), S : Total area of the surfaces which exhale radon (m^2), V : The volume of the room (m^3), C_{out} : Radon concentration outdoors (Bq m^{-3}), u : The ventilation rate of the room (h^{-1}), λ: Radon decay constant (h^{-1}), C_o: The radon concentration indoors (Bq m^{-3}).

From relationships (8) and (9)

$$C_o = \frac{C_{out} \cdot u - 2 \cdot S \cdot A \cdot \varepsilon \cdot \lambda \cdot l \cdot \sinh\frac{d}{2 \cdot l}}{\lambda + u} \quad (10)$$

4 Results and risk assessment

The results of the measurements are presented in the Table 1 and Table 2.

Table 1

CONCRETE

		Specific activities		Radon specific	Emanation
		$BqKg^{-1}$		exhalation rate	coefficient
			$BqKg^{-1}h^{-1}$	%	
	^{226}Ra	^{232}Th	^{40}K		
Range	7-41	1.2-4.7	57-96	0.004-0.024	5-25
Average	14	3.2	70	0.012	13
SD	7.7	0.8	10.3	0.004	4.6

Table 2

BRICKS

		Specific activities		Radon specific	Emanation
		$BqKg^{-1}$		exhalation rate	coefficient
			$BqKg^{-1}h^{-1}$	%	
	^{226}Ra	^{232}Th	^{40}K		
Range	25-83	35-65	539-1058	0.0013-0.035	0.5-12.4
Average	36	51.5	732	0.008	2.8
SD	12	8.9	158	0.009	3.2

(Savidou et al. 1995, Savidou et al. 1996)

4.1 Risk assessment from external irradiation

A unifying measure for materials containing different amounts of radium, thorium and potassium is the radium equivalent activity and is defined as :

$$Ra_{eq} = A(Ra) + 1.43\ A(Th) + 0.077\ A(K) \quad (11)$$

where A(Ra), A(Th) and A(K) are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K respectively, in $Bq \cdot Kg^{-1}$. This formula (Beretka and Mathew 1985) is based on the estimation that 370 $Bq \cdot Kg^{-1}$ ^{226}Ra, 259 $Bq \cdot Kg^{-1}$ ^{232}Th or 4815 $Bq \cdot Kg^{-1}$ ^{40}K result in the same gamma-ray dose rate (Stranden E. 1976, Krisiuk E. M. et al. 1971). To limit the radiation dose from building materials to

50 mrad/yr (ICRP77), many models have been suggested. One of the acceptable models, developed by (Krisiuk et al. 1971), leads to the inequality:

$$\text{Raeq} \leq 370 \ (12)$$

the higher radium equivalent activity (Raeq) in concrete and bricks used in Attica is 53 $Bq{\cdot}Kg^{-1}$ and 250 $Bq{\cdot}Kg^{-1}$ respectively which are well bellow the limit.

4.2 Risk assessment from the inhalation of radon and its daughters

The factors that influence the radon exhalation rate are: a) Ra-226 concentration in the building materials of Attica region in Greece Ra-226 concentration in concrete is from 15 to 55 ($Bq{\cdot}Kg^{-1}$) and in bricks it is from 120 to 260 ($Bq{\cdot}Kg^{-1}$) (Savidou A. et. al. 1995, 1996), b) Porosity: The porosity of building material varies from 0.05 to 0.25 (Culot M. V. J. et. al. 1976, Folkerts K. et. al. 1984), c) Radon emanation coefficient: The radon emanation coefficient for concrete varies from 0.05 to 0.25 and for bricks from 0.005 to 0.124 (Savidou A. et. al. 1995, 1996).

An indicative assessment of the risk from the inhalation of radon and its daughters in a typical room (4m x 3m x 2.7m) made of concrete is given here. The wall thickness is 0.2 m. For ^{226}Ra concentration in the walls C_{Ra}= 53 $Bq{\cdot}Kg^{-1}$ (the highest measured value) and exhalation coefficient is n=0.25 (the highest value) the radon concentration in the typical room air is 100 $Bq{\cdot}m^{-3}$. The density, the porosity, the diffusion length of the concrete and the radon concentration outdoors are considered to be 2400 kg m^{-3}, 0.25, 0.13 m and 1 $Bq{\cdot}m^{-3}$ respectively (UNSCEAR 88). The annual effective dose equivalent Heff (nSv) for ^{222}Rn is calculated as (UNSCEAR 88)

$$H_{eff} = 0.18 \cdot X_{Por,air} \cdot 8760(h \cdot \alpha^{-1}) \cdot 0.8 \ (13)$$

where 0.18 is a constant that refers to ^{222}Rn, X_{Rn-222} is ^{222}Rn concentration ($Bq{\cdot}m^{-3}$), 8760 are the hours per year and 0.8 is the mean breathing rate. The annual effective dose equivalent Heff (nSv) for the short-lived decay products of radon is calculated as (UNSCEAR 88)

$$H_{eff} = 9 \cdot X_{eqRn} \cdot 8760(h \cdot \alpha^{-1}) \cdot 0.8 \ (14)$$

where 9 is a constant that refers to the short-lived decay products of radon, X_{eqRn} is the equilibrium equivalent radon concentration ($Bq{\cdot}m^{-3}$) that is defined as (UNSCEAR 88)

$$X_{eqPor} = 0.5 \cdot X_{Por,air} \ (15)$$

Using equations (13), (14) and (15) we evaluated the annual effective dose equivalent for radon and its short-lived decay products to be 1600 μSv. We considered the maximum radon concentration in a room of concrete. We also considered the mean ventilation rate in Greece to be 1 h^{-1}. The estimated per caput effective dose equivalent from radon in areas of normal background is estimated to be 1100 μSv (UNSCEAR 88).

5 Conclusions

The higher radium equivalent activity (Ra_{eq}) in concrete and bricks used in Attica are well bellow the limit. The annual effective dose equivalent for radon and its short-lived decay products indoors, as it was evaluated for maximum radon concentrations indoor in Attica region is almost equal with the effective dose equivalent from radon in areas of normal background.

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