Estimation of Indoor Radon-222 Concentration in Homes from Measurements of Natural Exposure Rates (The MARNA Project)

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ABSTRACT

The aim of this study is to establish by means of a simplified model the mathematical expressions that relate external exposure rates due to natural gamma radiation in Spain to the potential concentrations of radon-222 gas inside buildings used as homes.

INTRODUCTION

The MARNA Project, being developed into the framework of an agreement subscribed between the Spanish Nuclear Safety Council (CSN) and the National Uranium Company (ENUSA), consists of four phases, the first three of them being already finished successfully in late December, 2000. The final objective of this Project was the elaboration of the natural gamma radiation map of Spain on the scale 1:1.000.000 by using radiometric data generated in the thirty years of lifetime of the ancient National Uranium Exploration and Investigation Plan, mainly through airborne, car-borne and by foot surveys, and those obtained within the MARNA Project itself. At the present, four other pilot maps on the scale 1:50.000 accomplished for four radiometric zones of special interest and three more on the scale 1:200.000 for the Autonomous Communities of Extremadura, Castilla-Leon and Galicia are also available. The research activities above mentioned have been carried out by two different research groups. In this paper, a simple mathematical model is shown to evaluate the indoor radon concentrations in homes from the data obtained in the MARNA project. The results achieved let us a good characterization of the affected areas in the country.

MATHEMATICAL MODEL

Suppose a solid is horizontally infinite and homogeneous: then, if the convective flow of a gas in a porous solid is ignored, the flow density of the gas (quantity of gas passing through the solid per unit of area and unit of time) is related to the concentration gradient of the gas in the pores of the solid according to Fick's law of diffusion:

$$J = - n D \ \delta C / \delta z \qquad \text{(Eq. 1)}$$

where

J: flow density of the gas (Bq m^{-2} s^{-1})

n: Corrected porosity of the medium: $n = (1 + w + kw)\ \varepsilon$

w: Water-filled fraction of the total pore volume of the medium

k: Solubility coefficient of radon in water

ε: Porosity of the medium

D: Effective diffusion coefficient of the gas in the solid (m^2 s^{-1})

C: Concentration of the gas in the air in the pores of the solid (Bq m^{-3})

z: Distance in the direction of diffusion (m)

For the case that concerns us, the solid is soil and the gas is radon (^{222}Rn).The ^{222}Rn concentration in the air in the pores of a stratum with a given ^{226}Ra content is determined by solving the diffusion equation

$$\delta C/\delta t = \delta/ \delta z\ (D\ \delta C/\delta z) - \lambda C + F \qquad \text{(Eq. 2)}$$

where

t: Time (s)

λ: Decay constant for ^{222}Rn (s^{-1})

F: Source term for ^{222}Rn in the air in the soil pores per unit of volume and unit of time (Bq m^{-3} s^{-1})

In the stationary state, the ^{222}Rn concentration is independent of time. In a first approximation, this can be considered to hold true for mean annual concentrations of the gas and, in this case, Eq. 2 can be simplified to take the form:

$$- \delta/\delta z\ (D\ \delta C/\delta z) + \lambda\ C = F \qquad \text{(Eq. 3)}$$

The rate of ^{222}Rn production in the air in soil pores depends on the ^{226}Ra concentration in the grains of material, since ^{222}Rn is produced by the radioactive decay of ^{226}Ra. The value of this rate of production can be calculated from the expression:

$$F = (E\ R\ \lambda\ \rho) / n \qquad \text{(Eq. 4)}$$

where

E: Emanation coefficient of ^{222}Rn (dimensionless)

ρ: Dry density of the soil (Kg m^{-3})

R: ^{226}Ra concentration in the soil (Bq kg^{-1}) of dry mass

The emanation coefficient E represents the ^{222}Rn fraction generated by ^{226}Ra decay that escapes from the solid soil particles and passes into the pores. This coefficient depends on many factors including the mineral composition of the soil, porosity, particle-size distribution and moisture content (proportion of water in the soil). Porosity ε is defined as the fraction of the total soil volume not occupied by solid particles.

To solve Eq. 3 the following boundary conditions are used:

- $C(z_a)=0$. The ^{222}Rn concentration in the air-soil interface is zero.
- $J(0)=0$. The ^{222}Rn flow is nil in the lowest part of the soil, so that the thickness of the soil layer under consideration, z_a, is assumed to be sufficient in relation to radon diffusion length (typically of the order of 10^{0} m).

The analytical solution for the surface exhalation rate of ^{222}Rn gas at its exit from the soil (z_a), when this is assumed to be homogeneous, is as follows:

$$J(z_a) = E\ R\ \rho\ (\lambda\ D)^{1/2} \tanh (z_a (\lambda/D)^{1/2}) \quad \text{(Eq. 5)}$$

If the depth of the soil is much greater than the ^{222}Rn diffusion length ($L= (D/\lambda)^{1/2}$) , then $\tanh (z_a (\lambda/D)^{1/2} \rightarrow 1$ and the following expression is obtained for the maximum surface exhalation rate of ^{222}Rn:

$$J_o = E\ R\ \rho\ (\lambda\ D)^{1/2} \quad \text{(Eq. 6)}$$

This expression relates the maximum amount of ^{222}Rn gas that can diffuse out of a soil containing a specific ^{226}Ra concentration. The ^{222}Rn concentration inside a home of base area A and of height H can be determined from the equation of mass conservation for an element of volume dV. In addition, this concentration depends on many variables including the characteristics of the building, geographical location, meteorological conditions, etc.

The variation of the mass of gas present in an element of volume dV is equal to the mass entering this element minus the mass leaving it. If the contribution from the building materials and from the ^{222}Rn concentration in the external atmosphere is ignored – and this is usually much lower than that present inside the building – the conservation equation in terms of activity can be approximated by means of the following mathematical equation:

$$\delta Q \;/\; \delta t \;=\; J_o \; A - (\lambda + v)\, Q \qquad \text{(Eq. 7)}$$

where

Q: Total ^{222}Rn activity inside the home (Bq)
A: Area of the base of the home (m^2)
v: Air renewal rate inside the home (s^{-1})

Bearing in mind that Q= C V, Eq. 7 can be written as:

$$\delta C \;/\; \delta t = \underbrace{J_o \; A/V}_{(1)} \; - \underbrace{(\lambda + v)\, C}_{(2)} \qquad \text{(Eq. 8)}$$

(1) Maximum entry rate of ^{222}Rn by diffusion from the soil per unit of volume of the home.
(2) Exit rate per unit of volume of the home.

In the stationary state, dC/dt=0 and, therefore, Eq. 8 is reduced to the expression:

$$C = \; J_o \;/\; (\lambda + v)\, H \qquad \text{(Eq. 9)}$$

Where

H: Height of the home (m)
C: ^{222}Rn concentration inside the building (Bq m^{-3})

There are three basic mechanisms responsible for air exchange between the external atmosphere and the inside of the building.

- Filtrations due to pressure gradients between the inside of the building and the external atmosphere through cracks, ceiling, floor and walls.
- Natural ventilation, that is, movement of air inwards or outwards caused by opening doors and windows.
- Forced mechanical ventilation, for example due to convection from air-conditioning installations.

Combining Eq. 6 and 9, we obtain a mathematical expression relating the ^{222}Rn concentration inside the home to the ^{226}Ra activity content per unit of soil mass. This expression is as follows:

$$C = \left(E\,R\,\rho\,(\lambda\,D)^{1/2}\right) / (\lambda + v)\,H \qquad \text{(Eq. 10)}$$

To relate the ^{222}Rn concentration to the external gamma exposure rate due to natural radiation sources obtained in MARNA, it is first necessary to calculate the exposure produced by an infinite homogeneous soil with 1 Bq kg^{-1} of ^{226}Ra at a height of 1 m above the ground. This calculation was performed with the *MICROSHIELD 5.05* code, and yielded the following relation:

$$T_{Ra\text{-}226} = 0.051\ R \qquad \text{(Eq. 11)}$$

where

$T_{Ra\text{-}226}$: Exposure rate at a height of 1 m above an infinite volume of homogeneous soil due to the content R (Bq kg^{-1}) of ^{226}Ra present in the soil (μR h^{-1})

0.051: Activity conversion factor per unit of mass of ^{226}Ra at exposure rate (μR h^{-1}Bq^{-1} kg).

The relation between the total γ exposure rate due to natural radiation and that due to ^{226}Ra has been determined experimentally (1) and the corresponding equation is as follows:

$$T_{Ra\text{-}226} = 0.2703\ T_{total} - 0.1219\ R \qquad \text{(Eq. 12)}$$

where T_{total} is the total γ exposure rate at a height of 1 m above the ground due to natural radiation (μR h^{-1}).

From Eq. 10, 11 and 12, we obtain the following empirical equation relating the indoor ^{222}Rn concentration in homes with the γ exposure rate due to natural radiation measured at a height of 1 m above the ground:

$$T_{total} = (0.18868\ C\ H\ (\lambda + v)) / (\ E\ \rho\ (\lambda\ D)^{1/2}\ ) + 0.45098 \qquad \text{(Eq. 13)}$$

If it is borne in mind that exposure to 1 Roentgen is equivalent to an absorbed dose in air of 8.7 mGy, Eq. 13 can also be expressed as follows:

$$S = 1.64152\ (C\ H\ (\lambda + v)) / (E\ \rho\ (\lambda\ D)^{1/2}) + 3.92353 \qquad \text{(Eq. 14)}$$

where *S* is the absorbed dose rate in air at a height of 1 m above the ground due to external gamma radiation expressed in (nGy h^{-1}).

The values inserted into Eq. 13 and 14 to estimate rates of exposure and absorbed dose in air at a height of 1 m above the ground and their relation with indoor ^{222}Rn concentrations measured in homes are those presented below.

Emanation coefficient	0.25	Default value from RESRAD 6.2
Renewal rate of air (s^{-1})	$1.39 \cdot 10^{-4}$	Default value from RESRAD 6.2
Density of soil (kg m^{-3})	1600	Default value from R.G. 3.64 and RESRAD 6.2
Soil moisture (w)	0.05	Default value from RESRAD 6.2
Porosity	0.39623	Calculated according to R.G. 3.64 (2)
Diffusion coefficient (m^2 s^{-1})	$5.914 \cdot 10^{-6}$	Calculated according to R.G. 3.64
Decay constant of ^{222}Rn (s^{-1})	$2.10 \cdot 10^{-6}$	
Surface area of home (m^2)	100	Default value from RESRAD 6.2
Height of home (m)	2.5	Default value from RESRAD 6.2
Activity conversion factor for ^{226}Ra at exposure rate (μR h^{-1} Bq^{-1} kg)	0.051	Obtained from MICROSHIELD 5.05

The equations used to calculate porosity, ε , and the effective diffusion coefficient of ^{222}Rn, D, are as follows:

$$\varepsilon = 1 - (\rho / 2.65 \rho_a) \qquad D = 7.10^{-6} \exp(-4(w - w\varepsilon^2 + w^5))$$

where

ρ_a: Density of water (1000 kg m^{-3})

With regard to the porosity value, the calculated value coincides with the default value used en the *RESRAD 6.2* code (3) and differs from that indicated in the document RP-107 (4), since the density of the soil used in this document is 2000 kg m^{-3}, which corresponds to a porosity of 0.25.

The value calculated for the effective diffusion coefficient is in good agreement with the default value assigned in the document RP-107 (4) of $5.0 \cdot 10^{-6}$ m^2 s^{-1} and can, therefore, be considered a fairly conservative value.

RESULTS AND CONCLUSION

Taking the above parametric values and assuming indoor ^{222}Rn concentrations of 150, 200 and 400 Bq m^{-3}, the results obtained from (Eq. 6, 10, 11, 13 and 14) are shown in Table I.

Indoor ^{222}Rn concentration	**150**	**200**	**400**
Maximum surface exhalation rate of ^{222}Rn (Bq m^{-2} s^{-1})	$5.29 \cdot 10^{-2}$	$7.05 \cdot 10^{-2}$	$1.41 \cdot 10^{-1}$
Specific activity for ^{226}Ra	$3.75 \cdot 10^{1}$	$5.00 \cdot 10^{1}$	$1.00 \cdot 10^{2}$
External gamma exposure rate due to ^{226}Ra (μR h^{-1})	1.91	2.55	5.10
Exposure rate due to external gamma radiation from the soil (μR h^{-1})	7.53	9.89	19.32
Absorbed dose rate in air due to external gamma radiation from the soil (nGy h^{-1})	65.51	86.04	168.08

Table I

For the results reflected in Table II, the classification of different geographical areas according to the potential risk associated with the presence of ^{222}Rn inside newly constructed homes would be as follows.

Type of Risk Area C(Bq m^{-3})	**Exposure rate due to natural external gamma radiation (μR h^{-1})**	**Absorbed dose rate in air due to external gamma radiation (nGy h^{-1})**
Low (C< 200)	< 10.3	< 90
Moderate (200≤C≤400)	10.3 – 18.4	90 – 160
High (C>400)	> 18.4	> 160

Table II

From these values, the potential radon map for the whole country is shown in figure 1. Of course, this process does not take into account the entry of radon gas due to transport phenomena caused by pressure differences between the inside of the home and the soil below it. In this regard, the Nuclear Safety Council has recently approved a research project (5) to be carried out by the University of Cantabria that will allow us to establish a better criterion on this issue.

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Figure 1