Applied Radiation and Isotopes 67 (2009) 849–853

A comparison between active and passive techniques for measurements of radon emanation factors

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ABSTRACT

Some radon related parameters have been determined through two different techniques (passive and active) in soil and phosphogypsum samples. Emanation factors determined through these techniques show a good agreement for soil samples while for phosphogympsum samples appear large discrepancies. In this paper, these discrepancies are analyzed and explained if non-controlled radon leakages in the passive technique are taken into account.

1. Introduction

Radon and its short-lived decay products are the most important contributors to human exposure to ionizing radiation from natural sources. This contribution represents 50% of the total dose (UNSCEAR, 2000) This significant level of radon radiology implies the need to control it in the workplace and/or housing. To do that, it is important to evaluate the role and contribution of the different materials that can act as radon sources inside buildings for work and residence. A common classification of materials based on the potential risk to radon exposure could be established based on the corresponding material exhalation rate. Nevertheless, the exhalation rate is a function of diffusion length, which, in turn, depends on several physical parameters of the material (humidity, porosity and geometry) and also depends on the methodology used for its determination (Petropoulos et al., 2001; Tuccimei et al., 2006). An alternative approach to quantify the potential risk due to the materials could be based on the radon potential, $\Omega = \varepsilon C_{Ra}$ (Bq kg^{-1}), being ε (dimensionless) the radon emanation factor, i.e., the fraction of the ^{222}Rn which has been produced by the decay of ^{226}Ra in a sample that is exhalated and C_{Ra} (Bq kg^{-1}) the ^{226}Ra activity concentration in the sample. Ω is also called the effective radium concentration because is the fraction of total radium which contributes to radon exhalation (Bossew, 2003). The radon potential is a characteristic of the material that is not influenced by the experimental conditions during its determination. The radon potential appears, then, as a suitable magnitude for classifying/comparing porous materials in relation to its potential radon radiological risk.

In this work, two techniques based on the determination of the exhalation rate within an accumulation chamber have been used to calculate the ^{222}Rn exhalation rates of different materials and their emanation factors: an active and a passive technique. The main differences between them are the time of accumulation and the evaluation of ^{222}Rn leakage through the chamber, which is included in the active technique and not considered in the passive one. Determination of ^{222}Rn emanation factor through passive methods based on accumulation techniques present several drawbacks as it has been described elsewhere (López-Coto et al., 2009 and references therein).

The main objective of this work is to emphasize the need of control the leakages in the sealed chamber technique. It would be desirable to obtain a correction factor for the passive technique in order to correct the measurements in a straightforward way. Nevertheless, this correction factor depends on the particular experimental conditions so it has to be determined for the particular measurement situation. Prior to ^{222}Rn related parameters measurement through a sealed chamber technique it is necessary to evaluate possible leakages by means of reference samples or intercomparison exercises.

2. Theoretical fundaments

The study of the accumulation of radon exhaled by a sample into a sealed chamber can be divided into two interdependent processes: (1) transport within the sample and (2) accumulation in the chamber after crossing the interface. The continuity condition of the radon concentration in the sample-air interface is the term relating both processes. These equations can be solved in an analytical way under several hypotheses.

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0969-8043/$ - see front matter doi:10.1016/j.apradiso.2009.01.045

2.1. Radon transport inside the sample

The generation and transport processes within porous materials can be described in a general way by an equation of balance for each phase in the sample (soil, water and interstitial air). The complexity of the problem can be reduced through a transformation of these equations into another for a simple phase (air), using expanded definitions of transport coefficients and porosity (Rogers and Nielson, 1991). This equation can be solved analytically under the following assumptions: One-dimension transport across z-axis; homogeneity of the diffusion coefficient and ^{226}Ra concentration in the sample; only diffusive transport happens (the pressure is constant within the chamber); Rn exhalation only happens at the sample-air interface ($z=0$). Under these conditions, the transport equation can be written as

$$\frac{\partial C_{Rn}}{\partial t} = \lambda_{Rn} \left. \quad l_0^2 \cdot \frac{\partial^2 C_{Rn}}{\partial z^2} + \Phi_0 - C_{Rn} \right) \tag{1}$$

$$C_{Rn}(z=0) = C_{Rn}^a \tag{2}$$

$$\left. \frac{\partial C_{Rn}}{\partial z} \right|_{z=-z_0} = 0 \tag{3}$$

where C_{Rn} is the radon concentration in the air-filled pores (Bq m^{-3}), l_0 the Rn diffusion length (m), z_0 the sample thickness (m), λ_{Rn} the radon radioactive decay constant (s^{-1}), Φ_0 is the source term of radon in the material (Bq m^{-3}) which is given by the equation:

$$\Phi_0 = \frac{C_{Ra} \cdot \rho \cdot \varepsilon}{\beta} = \frac{\rho}{\beta} \Omega \tag{4}$$

where β is the effective porosity, ρ the bulk density (g cm^{-3}), ε the emanation factor, C_{Ra} is the ^{226}Ra concentration in the sample (Bq g^{-1})

A detailed discussion of the terms and solutions of Eq. (1) can be seen elsewhere López-Coto et al., 2009. In this work, only the solution for particular condition has been described. Under stationary conditions, the solution of Eq. (1) leads to the exhalation rate as follows:

$$E = -l_0^2 \lambda_{Rn} \beta \left. \frac{\partial C_{Rn}}{\partial z} \right|_{z=0} = l_0 \lambda_{Rn} \beta (\Phi_0 - C_{Rn}^a) tgh\left(\frac{z_0}{l_0}\right) = E_0 - \omega C_{Rn}^a \tag{5}$$

where

$$\omega = l_0 \lambda_{Rn} \beta \, tgh\left(\frac{z_0}{l_0}\right) \tag{6}$$

E_0, the free exhalation rate (Bq m^{-2} s^{-1}), is obtained through the following equation:

$$E_0 = \omega \Phi_0 = l_0 \lambda_{Rn} \rho \, tgh\left(\frac{z_0}{l_0}\right) \Omega \tag{7}$$

In our case, the diffusion length is higher enough than the sample thickness ($l_0 \gg z_0$), in this way, a particular solution of the equation can be obtained as follows:

$$E_0 = \varepsilon C_{Ra} \rho_b \lambda_{Rn} z_0 = \lambda_{Rn} \Omega \frac{m}{S} \tag{8}$$

where S is the exhalation surface (m^2) and m is the sample mass (kg)

As the Rn exhalation rate measurements are performed in cylindrical geometry, the volume of the sample is given by $V = S\, z_0$.

2.2. Accumulation inside the chamber

The accumulation inside the chamber can be written as follows:

$$\frac{dC_{Rn}^a}{dt} = \frac{E_0 \cdot S}{V_c} - \lambda_{eff} C_{Rn}^a \tag{9}$$

where

$$\lambda_{eff} = \lambda_{Rn} + \lambda_b + \lambda_v$$

V_c is the effective volume of the chamber available for Rn accumulation, λ_v the chamber leakage time constant (s^{-1}), λ_b the bound exhalation time constant (s^{-1}), S the surface available for exhalation (m^2).

The solution of this equation assuming homogenous radon concentration inside the chamber can be expressed as

$$C_{Rn}^a(t) = C_{Rn}^0 \cdot e^{-\lambda_{eff} t} + \frac{E_0 S}{\lambda_{eff} V}(1 - e^{-\lambda_{eff} t}) \tag{10}$$

where C_{Rn}^0 is the Initial background concentration (Bq m^{-3}).

These sets of equations here described are valid for both the active and passive techniques. The main difference between them is that the active technique allows a continuum monitoring of the Rn concentration inside the chamber. In this way, an estimation of the leakage constant can be obtained from experimental results (López-Coto et al., 2007). Furthermore, the Rn exhalation rate and the Rn potential can be obtained in a shorter time by means of the active technique since it is possible to infer them from the initial slope of the Rn accumulation curve (Eq. (10)) inside the chamber (López-Coto et al., 2009). The active technique has been tested through different intercomparison exercises (López-Coto et al., 2007).

In the case of the passive method, Eq. (10) is still valid but only considering radioactive decay, i.e., $\lambda_{eff} = \lambda_{Rn}$. The only change that it is necessary to take into account is that initial Rn concentration in the can is negligible. In this way, the Rn concentration in the can at time t is given by

$$C(t) = \frac{E_0 S}{\lambda V}(1 - e^{-\lambda t}) \tag{11}$$

where S is the area covered by the can (m^2) and V the effective volume of the can (m^3).

The integrated radon activity after a period of time T as measured by the track detector is obtained through the expression:

$$I = \int_0^T C(t)\, dt \tag{12}$$

Then, the exhalation rate can be deduced from the following equation:

$$E_0 = \frac{I \lambda V}{S\left(T + \frac{1}{\lambda}(e^{-\lambda t} - 1)\right)} \tag{13}$$

3. Materials and methods

3.1. Samples

The soil and phosphogypsum samples were recorded from an industrial wastes disposal site, which has suffered different restoration processes since the beginning of its activity (see Bolívar et al., 2000, 2002 for details). The radiological state of the gypsum stacks and their surroundings after the different restoration steps has been evaluated in previous works (Mas, 2001; Mas et al., 2006).

3.2. Gamma-ray spectrometry

Gamma-ray measurements were performed using an extended range (XtRa) coaxial HPGe detector (Canberra), with 38% relative efficiency and resolution (FWHM) of 0.95 keV at the 122 keV line of ^{57}Co and 1.9 keV at the 1333 keV line of ^{60}Co. The detector was attached to a conventional electronic chain, including a multichannel analyzer, and was shielded with Fe 15 cm thick. ^{226}Ra activities were determined via the 352 keV emission of ^{214}Pb. This energy was chosen because the high emission probability (35.1(4)%) and the negligible summing effects. The whole procedure of calibration and gamma spectrometry measurements can be seen elsewhere (Pérez-moreno et al., 2002).

3.3. Radon related parameters measurements

3.3.1. Passive method

Can technique was used for the ^{222}Rn exhalation rate measurements through the passive method. A LR-115 type-II plastic track detector (2 cm × 2 cm) was fixed at the top inside of a cylindrical can made of High Grade Plastic having 7.5 cm height, 7.0 cm diameter and 0.5 mm thickness. A 100 g aliquot of each sample ($<100\,\mu$m grain size) was placed at the base of the can. The cans were sealed for 90 d. The sensitive lower surface of the detector is freely exposed to the radon produced by free emanation from the sample within the can. So that it could record alpha particles resulting from the decay of radon in the remaining volume of the can and from ^{218}Po and ^{214}Po deposited onto its inner walls. Radon and its daughter reach equilibrium in about 4 h and hence the equilibrium activity of emergent radon could be obtained from the geometry of the can and the time of exposure.

After the exposure the detectors were etched in 2.5 M NaOH at $(60\pm1)\,^{\circ}$C for a period of 90 min within a constant temperature water bath for detection of tracks. The resulting alpha tracks on the exposed face of the track detector were counted using an optical microscope at a magnification of 400 ×. The radon activity inside the can was obtained from the following expression:

$$C_{Rn} = kN_{tracks}$$

where C_{Rn} is the Rn activity inside the can (Bq m^{-3}), N_{tracks} the number of tracks on the detector and $k=(56.0\pm1.1)\times10^{-3}\,\mathrm{cm^{-2}\,d^{-1}\,(Bq\,m^{-3})^{-1}}$ is a calibration factor obtained from an earlier calibration experiment (Singh et al., 1997). Details of the whole procedure can be seen elsewhere (Mahur et al., 2008).

3.3.2. Active method

Experimental measurements were performed with a radon accumulation chamber made with a cylindrical sample-holder screwed in the radon detector and sealed with a rubber gasket. The accumulation volume was around 0.6 L. For radon concentration monitoring a radon detector AlphaGUARD PQ2000PRO of Genitron Instruments (AG) has been used. This is an ionization chamber which works in a 2.2×10^{6} Bq m^{-3} range and a temporal resolution of 10 min. This detector has been factory-calibrated and is traceable to PTB and NIST. The samples were prepared in cylindrical geometry and the sample thickness is always less than 5 cm. The walls of the sample container ensure that radon exhalation from the materials only appears on their upper surface. A detailed description of the experimental system and the whole procedure of calibration of the radon accumulation chamber can be seen elsewhere (López-Coto et al., 2007).

4. Results and discussion

The radon exhalation rate is influenced by the sample properties and experimental conditions. Both, the passive and active methods provide the exhalation rate as a prompt result. In order to determine a parameter independent on the experimental conditions, the emanation factor has been calculated for all the analyzed samples through Eq. (8).

Table 1 presents the ^{226}Ra activity concentrations and the emanation factors (ε) for soil and phosphogypsum samples. The emanation factor have been calculated for passive (ε_p) and active (ε_a) techniques.

As can be seen from this table, ^{226}Ra activity concentrations are in the range 11–35 Bq kg^{-1} for soil and 480–770 Bq kg^{-1} for phosphogypsum samples in agreement with typical values. The average of ^{226}Ra activity concentrations for soils and phosphogypsum samples are 22 and 624 Bq kg^{-1} respectively. From Table 1 it is also possible to infer discrepancies between emanation factors calculated through both techniques. If we take into account that the physical state of the samples and sample itself are the same in each method, the difference in emanation factor could be attributed on principle to systematic error between both techniques or to non-controlled ^{222}Rn leakages in passive technique.

Table 1 also lists the ratios between emanation factor calculated through the passive and active techniques. As it can be seen, these ratios range between 0.6 and 1.7 for soil samples while for phosphogypsum samples is one order magnitude lesser than in soil. Furthermore, the emanation factor calculated through the passive technique for phosphogypsum samples are underestimated in respect to those one calculates through active technique and to those published in literature (De Martino and Sabbarese, 1997; Ferry et al., 2002); on the contrary, in the case of soils samples the emanation factor from the passive technique is slightly overestimated. This fact discards the presence of systematic error between both techniques, otherwise the emanation factor ratios should be constant for all the samples.

Table 1
^{226}Ra activity concentrations (C_{Ra}) and emanation factors measured by passive (ε_p) and active (ε_a) techniques for soil samples (Table a) and phosphogypsum samples (Table b).

Sample code	C_{Ra} (Bq kg^{-1})	ε_p (%)	ε_a (%)	$\varepsilon_p/\varepsilon_a$
(a) Soil samples				
1-1-S	11.8 ± 0.5	45 ± 3	25.8 ± 1.8	1.74 ± 0.17
1-2-S	12.5 ± 0.6	49 ± 3	28.4 ± 1.9	1.73 ± 0.17
1-3-S	23.0 ± 1.2	24.1 ± 1.7	31.1 ± 2.2	0.77 ± 0.08
1-4-S	27.3 ± 1.6	29.5 ± 1.9	20.2 ± 1.3	1.46 ± 0.14
1-5-S	33.3 ± 1.6	20.4 ± 1.7	37 ± 3	0.55 ± 0.06
1-6-S	23.6 ± 1.3	23.9 ± 1.6	15.1 ± 1.0	1.58 ± 0.15
1-7-S	20.0 ± 0.9	25.9 ± 1.5	22.7 ± 1.3	1.14 ± 0.09
1-8-S	15.1 ± 0.6	32.3 ± 2.3	25.3 ± 1.8	1.28 ± 0.13
1-9-S	30.4 ± 2.1	24.2 ± 1.5	37.7 ± 2.3	0.64 ± 0.05
Average	22 ± 2	31 ± 3	27.0 ± 2.5	1.1 ± 0.5
(b) Phosphogypsum samples				
1-1-G1	590 ± 20	1.13 ± 0.05	21.3 ± 1.0	0.053 ± 0.003
1-1-G2	480 ± 20	2.34 ± 0.12	28.0 ± 1.4	0.084 ± 0.006
1-2-G1	650 ± 21	0.97 ± 0.05	15.2 ± 0.9	0.064 ± 0.005
1-2-G2	620 ± 25	0.61 ± 0.03	12.1 ± 0.6	0.050 ± 0.003
1-3-G1	580 ± 22	1.26 ± 0.05	19.0 ± 0.8	0.066 ± 0.004
1-3-G2	570 ± 16	1.98 ± 0.12	22.4 ± 1.3	0.088 ± 0.008
1-6-G1	730 ± 20	0.84 ± 0.06	16.8 ± 0.7	0.050 ± 0.004
1-6-G2	770 ± 30	0.94 ± 0.05	18.5 ± 1.1	0.051 ± 0.004
Average	624 ± 30	1.26 ± 0.21	19.2 ± 1.7	0.07 ± 0.04

The ratios between emanation factors calculated through both techniques are also shown. Uncertainties are quoted at one sigma level.

Let us assume then that discrepancies are due to non-controlled ^{222}Rn-leakages in the experimental system used in the passive setup. As the chambers where the samples are sealed for ^{222}Rn in growth are stored all together, it is possible to lead a situation in which the ^{222}Rn leakages from several chambers may act as source of ^{222}Rn for other ones. If a chamber is not perfectly sealed and there is a gradient of ^{222}Rn concentration between indoor and outdoor air, a flux of ^{222}Rn atoms through its boundary may be established. If outdoor ^{222}Rn concentration is higher than indoor concentration, the flux of ^{222}Rn atoms occurs towards the inner side of the chamber, so ^{222}Rn concentration increases within. On the contrary, if indoor ^{222}Rn concentration is higher than outdoor concentration, the flux of ^{222}Rn atoms goes outwards the chamber causing a decrease in the ^{222}Rn concentration.

As can be seen from Table 1, the ^{226}Ra activities obtained for phosphogypsum samples are higher than for soil samples. Therefore, it can be expected that ^{222}Rn concentration in the chambers with phosphogypsum samples be significantly higher than in chambers with soil samples. In this way, a net flux outwards of ^{222}Rn atoms can be expected for the chambers with phosphogypsum samples leading to underestimation of ^{222}Rn exhalation rates, as it happens with the emanation factor. On the contrary, an overestimation of emanation factor can be expected for the soil samples. In this way, the leakages from the chambers having highest ^{222}Rn concentration (due to phosphogypsum) leads to an increase of the outdoor concentration and influences the measurements of the soil samples.

An apparent leakage constant for the passive technique can be estimated based on the following considerations: after a period of ^{222}Rn in-growth, the ^{222}Rn saturation concentration in the "can" is given by

$$C_{Rn}^{Sat} = \frac{E_0 S}{\lambda_{eff} V} \tag{14}$$

where E_0 is the ^{222}Rn exhalation rate (Bq m^{-2} h^{-1}), S the surface available for exhalation (m^2), V the volume of the can, λ_{eff} the same meaning as in Eq. (9).

In the can technique, the ^{222}Rn saturation concentration is estimated through the equation:

$$C_{Rn}^{Sat} = \frac{E_{ap} S}{\lambda_{Rn} V} \tag{15}$$

E_{ap} is the apparent ^{222}Rn exhalation rate (Bq m^{-2} h^{-1}). ^{222}Rn leakages are neglected, λ_{Rn}: ^{222}Rn radioactive decay constant.

S and V have the same meaning than in preceding equation.

Taking into account that C_{Rn}^{Sat} is an experimental data obtained in each situation, considering or not ^{222}Rn leakages, it is possible to infer:

$$\frac{E_0}{\lambda_{eff}} = \frac{E_{ap}}{\lambda_{Rn}} \tag{16}$$

E_{ap} is obtained through the ^{222}Rn emanation factor neglecting leakages, ε_p. On the other hand, as the ^{222}Rn emanation factor is a property of the material, it does not depend on the experimental set up used to calculate it. In this way, it is possible to assume that ^{222}Rn emanation factor obtained through the active technique, ε_a, is the true value. Therefore, as the diffusion length is higher enough than samples thickness, the ^{222}Rn exhalation rate can be written as

$$E_0 = \lambda_{Rn} \varepsilon_a C_{Ra} \frac{m}{S} \tag{17}$$

$$E_{ap} = \lambda_{Rn} \varepsilon_p C_{Ra} \frac{m}{S} \tag{18}$$

C_{Ra} is the ^{226}Ra activity concentration in the sample (Bq kg^{-1}) and m the sample mass (kg).

Therefore, a relationship between both ^{222}Rn emanation factors, true and apparent, can be obtained:

$$\lambda_{eff} = \frac{\varepsilon_a}{\varepsilon_p} \lambda_{Rn} \tag{19}$$

As consequence, the ^{222}Rn leakage constant for the passive technique can be deduced as

$$\lambda_v^{apparent} = \left(\frac{\varepsilon_a}{\varepsilon_p} - 1 \right) \lambda_{Rn} - \lambda_b \tag{20}$$

Table 2 presents the apparent leakage constant obtained in the measurement of ^{222}Rn-related parameters through the passive technique.

As can be seen from this table the leakage constant for soils are negative excepting samples 1-3-S, 1-5-S and 1-9-S. This fact indicates that in the case of soils there is a very slight net flux of ^{222}Rn atoms inwards. On the other hand, in the case of phosphogypsum samples all the leakage constants are positive. This fact implies a net flux of ^{222}Rn atoms outwards. The leakage constants for phosphogypsum samples are two order magnitude higher than for soils, as can be expected according to their higher ^{226}Ra activity. The absolute values of ^{222}Rn leakage constant for soils are one magnitude order lesser than ^{222}Rn radioactive decay constant (2.1×10^{-6} s^{-1}) while for phosphogypsum samples are one order magnitude higher. These values show that ^{222}Rn leakage is a significant contributing factor that it is necessary to take into account in order to estimate ^{222}Rn related parameters through the accumulation chamber technique. On principle, and taking into account that ^{226}Ra activity concentrations are significant lesser in soils than in phosphogypsum samples, it could be expected a negative leakage constant for all the soils samples. Nevertheless, as ^{222}Rn emanation factor can vary from sample to sample, the magnitude of leakage constant will depend on the ^{222}Rn potential ($\Omega = \varepsilon C_{Ra}$(Bq kg^{-1})) and not only on the ^{226}Ra activity concentration in the sample.

Fig. 1 shows the dependence of the chambers leakages constants on the radon potential of the samples obtained through a weighted linear fitting by least square method. The ^{222}Rn leakage constant for soils samples show a strong lineal dependence on the radon potential ($r = 0.91$). This fact suggests that the leakage constant can be related to the difference between radon external concentration and the concentration inside the measurement chamber. As can be seen from this figure, the functional relationship between ^{222}Rn leakage constant and radon potential is the same when considering soil samples and all the samples. This fact indicates that the more the radon potential is, the higher the ^{222}Rn leakage effect is. The slope and intercept in both cases are significant at 0.001 level.

Table 2
^{222}Rn leakage constant in the measurement of each sample through the passive technique.

Sample code	λ_v (10^{-7} s^{-1})	Sample code	λ_v (10^{-5} s^{-1})
1-1-S	-8.3 ± 1.2	1-1-G1	3.9 ± 0.3
1-2-S	-8.3 ± 1.2	1-1-G2	2.4 ± 0.2
1-3-S	8 ± 3	1-2-G1	3.3 ± 0.3
1-4-S	-5.9 ± 1.3	1-2-G2	4.2 ± 0.3
1-5-S	19 ± 4	1-3-G1	3.1 ± 0.2
1-6-S	-7.1 ± 1.3	1-3-G2	2.3 ± 0.2
1-7-S	-1.7 ± 1.5	1-6-G1	4.2 ± 0.4
1-8-S	-3.8 ± 1.6	1-6-G2	4.1 ± 0.3
1-9-S	13 ± 3		

Errors are quoted at one sigma level.

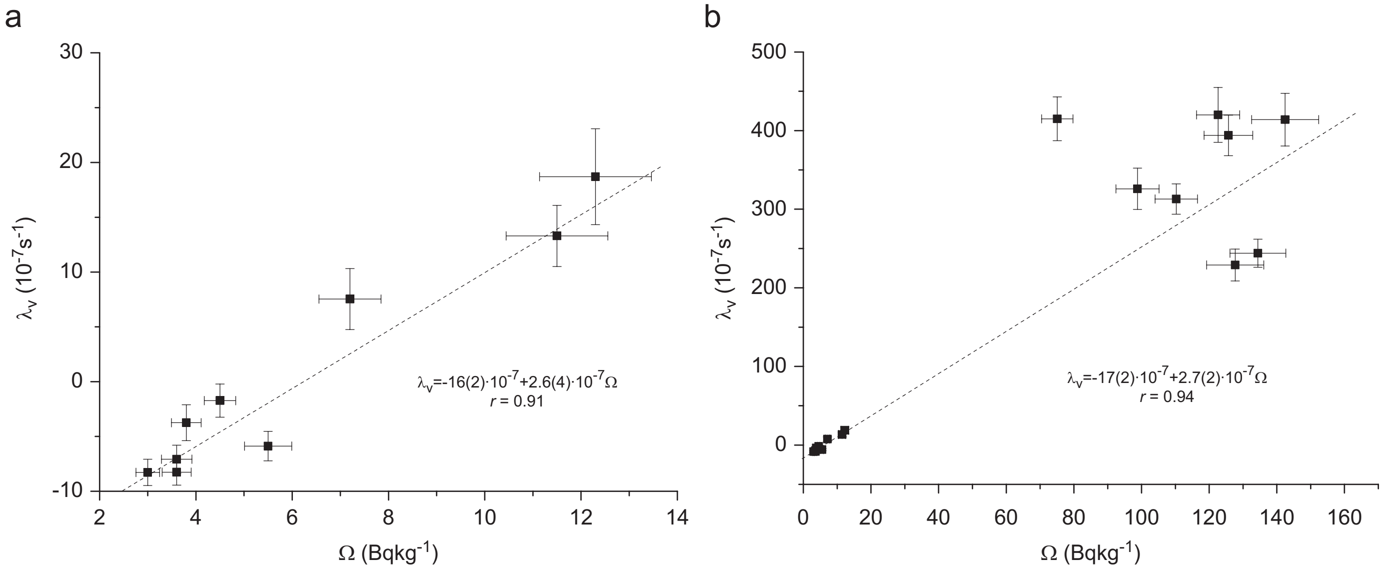

Fig. 1. Leakages constant as function of radon potential of the samples. Number within parentheses refer to the error of the last figure: (a) soil and (b) all samples.

These results are suggesting the need to revise the leakages model. Until now, the model has been related only with the inside radon concentration. This approximation could be accepted when the outdoor concentration is very lower than that inside of the chamber. Nevertheless, in this experiment the outdoor concentration played a very important role in the net leakages showed in each chamber.

5. Summary and conclusions

The ^{222}Rn emanation factors have been calculated for soil and phosphogypsum samples from an area where the wastes of phosphate rock processing plants are stored. The ^{222}Rn emanation factor has been calculated through an active and a passive technique. The results show discrepancies that can be explained by the presence of uncontrolled ^{222}Rn leakages in the passive technique. These discrepancies have been used to calculate an apparent leakage constant for each of the chamber used in the passive technique. The results indicate the need for controlling the leakages when measuring ^{222}Rn-related parameters as exhalation rates or emanation factor.

The leakage correction factor for the passive technique is a characteristic of the experimental system and the particular conditions used in the measurements of Rn related parameters. The determination of this correction factor requires an intercomparison with some well established technique or to work with references samples with known ^{222}Rn related parameters. The correction factor depends on the ^{222}Rn concentration in the storage room which in turns depends on the dimensions of the room, the number of samples simultaneously and the Rn potential of the whole set of samples.

Acknowledgments

This work has been supported by the Spanish Department for Science and Technology through the research projects REN2003-04942, CTM2006-08148/MAR, the Spanish Nuclear Council Authority project: "Radiological Impact Produced by Chemical Industries Located in the South of Spain" and the Regional Government of Andalusia.

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