Ann. Geophysicae 15, 124–133 (1997)

Annales Geophysicae

Release of ^{222}Rn from some soils

C. Dueñas, M. C. Fernández, J. Carretero, E. Liger, M. Pérez

Department of Applied Physics, Faculty of Sciences, University of Málaga, 29071 Málaga, Spain

Received: 25 October 1995/Revised: 14 June 1996/Accepted: 3 July 1996

Abstract. Measurements have been made of ^{222}Rn release from diverse soils in the region surrounding Málaga, Spain. These flux measurements were carried out by two methods. A direct method using a static chamber technique and another indirect method obtained from concentration profile measurements of ^{222}Rn in the soil air. The effects of meteorological variables and other parameters on ^{222}Rn flux were studied. The factors that most affected the instantaneous value of ^{222}Rn release were humidity and soil thermal gradient. The directly measured ^{222}Rn fluxes at investigated sites are higher than ^{222}Rn fluxes derived by the indirect method.

1. Introduction

^{222}Rn is an inert radioactive element with a half-life of 3.8 days. It belongs to the radioactive uranium series and occurs in soil gas in varying concentrations. In recent years, ^{222}Rn has been used as a tracer for the origin and trajectory of air masses (Prospero and Carlson, 1970; Karol, 1974; Wilkniss *et al*., 1974; Larson and Bressan, 1980). ^{222}Rn has also been used in more quantitative studies particularly to determine vertical matter diffusion coefficients (Druilhet and Fontan, 1972; Hsu *et al*., 1980). Another interesting use of ^{222}Rn is as an indicator of the vertical stability of the lower atmosphere at a given site through a study of the equivalent mixing height, a parameter which is a characteristic of vertical stability (Guedalia *et al*., 1980). In problems relating to conservation of mass of atmospheric ^{222}Rn an accurate knowledge of ^{222}Rn release is essential.

The present research was undertaken with three principal goals: (1) to characterize the flux of ^{222}Rn from the soils of Málaga; (2) to study which factors are responsible for the variation in flux from one site to the next and (3) to compare the flux of ^{222}Rn by two different methods.

However, the ^{222}Rn release value is in itself an important parameter, as it is directly related to the presence or absence of seismic activity (King, 1978; Dueñas and Fernández, 1988) and also to the effects of underground nuclear explosions. However, the numerical value of ^{222}Rn release for a given terrain is not the only important factor. It is equally necessary to know the variations of the release that may be caused by changes in meterological conditions. The characteristics of the soil itself are important, including the amount of parent ^{226}Ra present, the porosity and permeability of soil and the degree of water saturation. There is a great confusion and a lot of argument about the results obtained, especially regarding the influence of certain meteorological parameters.

In the present work, the release of ^{222}Rn was measured by two methods, including closed accumulation and an indirect method based on an understanding of the vertical profile of the ^{222}Rn present in the soil air in the first few meters below the ground surface. The ^{222}Rn release has been measured in four types of soil in the area around Málaga city (Spain) over various times. Quantitative relations between the ^{222}Rn release and several meteorological variables and characteristics of the soil were found. ^{222}Rn transport in the unsaturated soil zone is by molecular diffusion. We have determined the diffusion coefficient and the emanation fraction for all the soils studied. Finally, the ^{222}Rn release measured by accumulation method is compared for each soil with the exhalation obtained indirectly from concentration profile measurements in the soil air.

2 Theoretical considerations

Gas transport in the unsaturated soil zone occurs through molecular diffusion; then, the ^{222}Rn flux can be calculated from Fick's first law:

$$\vec{J}_{Rn} = -D_{Rn}\vec{\nabla}C_{Rn}, \quad (1)$$

Correspondence to: C. Dueñas

where $\vec{J}_{Rn}$ is the flux density, D_{Rn} is the bulk diffusion coefficient for ^{222}Rn through the volume of the porous medium and C_{Rn} is the concentration of ^{222}Rn in the interstitial gas.

Fick's second law follows from Eq. (1) by a conservation principle with the added terms due to ^{222}Rn decay and emanation from the solids of the medium, assuming a homogeneous ^{222}Rn source strength and a depth independent diffusion coefficient D_{Rn}:

$$\frac{\partial C_{Rn}}{\partial t} = \frac{D_{Rn}}{\varepsilon} \nabla^2 C_{Rn} - \lambda_{Rn} C_{Rn} + \Phi_{Rn}, \tag{2}$$

where ε is the porosity of the medium defined as the ratio of void volume to total volume, λ_{Rn} is the ^{222}Rn decay constant and Φ_{Rn} is the emanation power of the medium into the interstitial volume. Φ_{Rn} (Bq m^{-3} s^{-1}) is estimated from soil parameters and the decay ^{222}Rn constant (Nazaroff *et al.*, 1988) according to:

$$\Phi_{Rn} = \frac{f \cdot \lambda_{Rn} \cdot \rho_d \cdot C_{Ra-226}}{\varepsilon}, \tag{3}$$

where f is the emanation fraction, defined as the quotient between the ^{222}Rn atoms that enters the pore soil and the ^{222}Rn atoms generated in soil, ρ_d is the density of soil grain and C_{Ra-226} (Bq kg^{-1}) is the radium activity concentration in the soil.

Equation (2) may be solved in the steady state using the boundary conditions $C_{Rn} \to 0$ at $z \to 0$ and $C_{Rn} = C_{\infty, Rn}$ at $z \to \infty$:

$$C_{Rn}(z) = C_{\infty, Rn}(1 - e^{-(z/\bar{z}_{Rn})}), \tag{4}$$

where:

$$\bar{z}_{Rn} = \sqrt{\frac{D_{Rn}}{\varepsilon \lambda_{Rn}}} \tag{5}$$

is a denominated relaxation depth and $C_{\infty, Rn}$ is the radioactive equilibrium concentration value at great depth.

From Eqs. (4), (5), (2) and (3), the equation of emanation fraction of ^{222}Rn is obtained:

$$f = \frac{\varepsilon \cdot C_{\infty, Rn}}{\rho_d \cdot C_{Ra-226}}. \tag{6}$$

This equation permits us to obtain the emanation fraction of ^{222}Rn from soil as a function of physical characteristics of soil, such as porosity, density and radium content and $C_{\infty, Rn}$.

The flux at the soil surface, $J_{0, Rn}$, as a function of relaxation depth is therefore:

$$J_{0, Rn} = -D_{Rn} C_{\infty, Rn} \sqrt{\frac{\varepsilon \lambda_{Rn}}{D_{Rn}}} = -\frac{D_{Rn} C_{\infty, Rn}}{\bar{z}_{Rn}}. \tag{7}$$

All the equations are derived under the assumption that the gas transport in the soil water phase can be neglected. This assumption is true for ^{222}Rn because the molecular diffusion coefficient of ^{222}Rn is 10^4 times smaller in water than in air. In Eq. (1) we have neglected the transport due to macroscopic flow because, assuming Darcy's law, the macroscopic flow (J_{mac}) is:

$$J_{mac} = C \cdot v = C \cdot \left(-\frac{K}{\eta} \frac{dp}{dz} \right),$$

where K is the permeability, η is the dynamic viscosity and p is the absolute pressure. In the study soils, the K values have been small. The soil properties will be exposed after this paragraph.

3 Material and methods

3.1 The chamber

The closed accumulation methods were of the type described by Wilkening *et al.* (1972). The chamber used consists of a cylindrical container 0.55 m in diameter and 0.32 m in height. The chamber was made of stainless steel with a sampling tube. A small electric fan inside the chamber was used to maintain uniform mixing of emitted gases. The fan was operated a few minutes before the accumulation period was finished. Equal pressure between the environment and the interior of the chamber was maintained through an orifice with 3 mm diameter. The chamber was placed at a depth of 1–2 cm in the soil.

3.2 Radium content

The radium content of the soil is typically given as an activity concentration per unit mass. Soil cores were obtained in the first centimeter of the soil. Samples were dried in the laboratory, sieved to remove rock fragments and sealed in Marinelli type container of 450 cm^3 for several weeks. Samples were counted directly on an intrinsic germanium detector for 24 h to obtain sufficient precision of measurement. The intrinsic germanium detector has an efficiency of 25%, a resolution of 2 KeV and is surrounded by shielding material to reduce the background count. The 610 KeV gamma of ^{214}Bi was used as a measurement of ^{226}Ra activity after several weeks had elapsed between sealing and gamma counting to permit ^{214}Bi to grow into equilibrium with ^{226}Ra. Careful calibration was carried out using soil samples of similar density with ^{152}Eu uniformly distributed and sealed in the Marinelli recipient analogues used for the soil cores.

3.3 Radon in air

For the ^{222}Rn analysis, air was sampled into a 1 l or 0.5 l lucite walled cylindrical cell, previously evacuated, as a modified design of the original Lucas cell (Lucas, 1957) developed by Quindós *et al.* (1991). The walls of the cell were coated internally with SZn(Ag) coated mylar, but the main difference from the traditional one is the ability to open the cell after use by removing the bottom. The background count of this kind of cell ranged from 0.7 to 1.3 c.p.m., with counting times of 30 min, resulting in

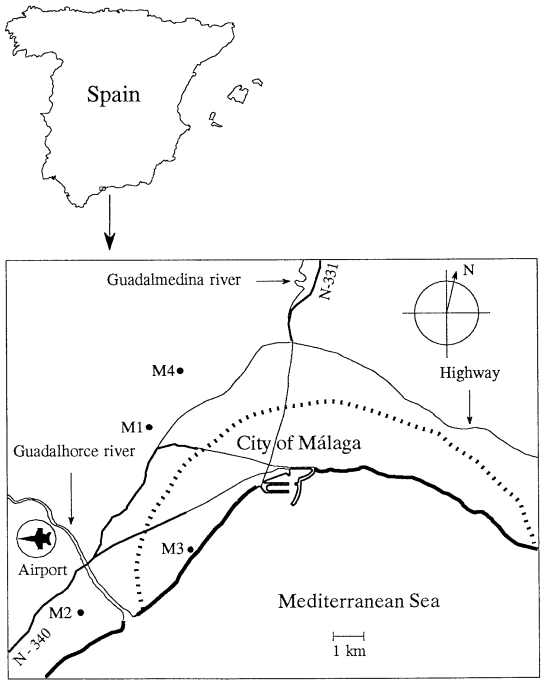

Fig. 1. The location of sampling points

a lower limit of detection around 10 Bq/m^3. ^{222}Rn was measured by counting the alpha particles emitted by ^{222}Rn and its daughter products, ^{218}Po and ^{214}Bi, when they reach radioactive equilibrium. The precision of the measurements was about 5% taking only the statistical error into account (Carretero, 1994). Typically, counting intervals of 30 min to 2 h were employed. The cell developed by Quindós *et al.* (1991), was calibrated in the Lovelace Inhalation Toxicology Research Institute, Albuquerque, New Mexico, USA. Afterwards the cell was recalibrated in our laboratory obtaining an average efficiency of 0.510 ± 0.015.

3.4 Interstitial ^{222}Rn air

Measurements of ^{222}Rn in soil air concentrations were made by inserting some stainless steel sampling tubes into the soil at several depths, according to the soil type. The 10 mm diameter tubes ended in a 3.5 cm diameter, 6 cm long *filtration chamber* which was designed to permit the aspiration of interstitial air from the soil. The chamber was filled with glass fibers to prevent the aspiration of soil or clay particles. The upper ends of the tubes were sealed except for the time that samples were being taken. A small purging took place before and after sampling was completed, by direct (filtered for dust) intake into evacuated scintillation cells for subsequent ^{222}Rn analysis.

3.5 Location and properties of soils

All the samples were taken in four sites in an area located outside the city of Málaga. The climate of Málaga is warm, temperate with hot summers and little rain, particularly during measurement periods in the last 2 years. Figure 1 shows the location of study soils and Table 1 exhibits the characteristics of the soils mentioned.

We studied soil properties such as particle-size analyses, density, porosity and permeability. Table 2a shows the grain size distribution and Table 2b the different properties.

Particle size analyses were carried out by the sieve method as in Jiménez and de Justo (1975). Porosity was determined in the laboratory from real and apparent density measurements of a soil sample obtained by perforation of the soil with a core sampler which is a hollow cylinder whose volume is known. Permeability was determined in situ assuming Darcy's law and measuring the flow for a fixed pressure drop.

The meteorological data (wind speed, air temperature, air humidity and atmospheric pressure) were supplied by the Málaga Airport Observatory. Soil temperatures at 15 cm and 80 cm depth were measured with a Crisson digital thermometer with a platinum sounding probe. Soil humidity was measured during all the samplings on the first centimetre of soil.

4 Results and discussions

4.1 The ^{222}Rn release by the direct method

Soil gas flux was measured directly by the accumulation method (Wilkening *et al.*, 1972) using the chamber described in Sect. 3.1. The concentration change was

Table 1. Characteristics of the sampling sites in Málaga

Characteristics	M1	M2	M3	M4
Site location	Semiurban	Semiurban	Urban	Semiurban
Vegetation type	Bare	Ficus and Eucalyptus	Bare	Pine
Hours of sunshine	All day	2 h Sunset	All day	Zero
Period of meaasurements	June 91 January 93	January 92 June 93	February 92 October 92	June 92 June 93
Average soil temperature at 15 cm depth	(19 ± 6) °C	(17 ± 4) °C	(23 ± 7) °C	(17 ± 5) °C
Average humidity over the first cm	(7 ± 6)%	(12 ± 6)%	(8 ± 6)%	(7 ± 5)%
^{226}Ra content (Bq/kg)	30.1 ± 1.0	39.9 ± 1.8	26.0 ± 1.0	24.8 ± 0.9

Table 2

	M1	M2	M3	M4
a. Grain size distribution				
Grain size				
Clay and slime	32%	88%	15%	21%
Fine sand	13%	4%	24%	26%
Coarse sand	55%	8%	61%	53%
	M1	M2	M3	MV4
b. Properties of the soils				
Properties				
Density (g/cm^3)	1.76 ± 0.02	1.58 ± 0.02	1.75 ± 0.02	1.52 ± 0.02
Porosity (%)	25.9 ± 0.3	42.9 ± 0.1	36.4 ± 0.4	43.4 ± 0.3
Permeability (ms)	$5.88 \cdot 10^{-7}$	$< 10^{-7}$	$2.32 \cdot 10^{-6}$	$3.72 \cdot 10^{-6}$
Soil type	Soft clay Slightly permeable	Resistant clay Impermeable	Compact sandy Moderately permeable	Loose sandy Moderately permeable

Table 3. Average ^{222}Rn flux, activity of ^{226}Ra and number of measurements for each type of soil

Location	M1	M2	M3	M4
$\bar{J}_{Rn} \cdot 10^{-3}$ (Bq m^{-2} s^{-1})	12 ± 8	25 ± 13	3.0 ± 1.6	10 ± 4
^{226}Ra (Bq kg^{-1})	30.1 ± 1.0	39.9 ± 1.8	26.0 ± 1.0	24.8 ± 0.9
Number of measurements	82	73	30	50

measured after an accumulation period of 30 min to 1 h. The flux density is calculated by:

$$J_{Rn} = h \frac{\Delta C_{Rn}}{\Delta t},$$

where ΔC_{Rn} is the concentration increase during Δt and h is the height of the chamber. The average accuracy of ^{222}Rn flux by direct method has been 7%. Table 3 shows the average flux of ^{222}Rn accompanied by an indication of the standard deviation found, the activity of ^{226}Ra and the total number of measurements for each soils.

Altogether, we obtained 235 individual ^{222}Rn release measurements for soils situated close to Málaga city. The range of variations of these values is between $3 \cdot 10^{-3}$ and $25 \cdot 10^{-3}$ Bq m^{-2} s^{-1}, which is coherent with the global distribution of ^{222}Rn release except for soil M3. Taking into account the different methods existing in order to determine the ^{222}Rn release in the interphase soil-atmosphere, ^{222}Rn release oscillates between 0.5 and 2.5 atoms cm^{-2} s^{-1} ($10 \cdot 10^{-3}$ and $52 \cdot 10^{-3}$ Bq m^{-2} s^{-1}), according to existing abundant literature on the measurement of ^{222}Rn release (Israël, 1951; Servant, 1964; Pearson and Jones, 1965; Wilkening *et al.*, 1972; Sisigina, 1967; Turekian *et al.*, 1977; Mochizuki and Sekikawa, 1978; Martín Salas *et al.*, 1979; Quindós, 1980; Lambert *et al.*, 1982; Nero and Nazaroff, 1983; Polian *et al.*, 1989 and Graustein and Turekian, 1990.

The sampling time was always at the same time of day (1500 UT). The average ^{222}Rn release from M3 soil is very low for two reasons: (a) their typical location on the coast (30 m from the sea shore) where the water table is about 1.20 m below the surface in dry periods; for this, the humidity at depth is high, the migration of ^{222}Rn towards the interphase soil-atmosphere making difficult and (b) the number of samplings when rains fell close to the sampling, being 50% of all measurements.

The ^{222}Rn release is related to the activity of ^{226}Ra for each soil. A dependence of ^{222}Rn release on the activity of ^{226}Ra was observed which may be expressed by the following equation:

$$\bar{J}_{Rn} = 1.2 \cdot 10^{-3 \cdot 226}\text{Ra} - 2.5 \cdot 10^{-3}$$

which gives a correlation coefficient of 0.93 and a degree of confidence higher than 90%.

According to Sisigina (1967) and Dörr and Münnich (1990) the ^{222}Rn release from soil may be influenced by the radium content and the grain size diameter of soil particles. This is confirmed by the observation of lower ^{222}Rn fluxes ($3 \cdot 10^{-3} - 10 \cdot 10^{-3}$ Bq m^{-2} s^{-1}) on sandy soils (M3 and M4) and higher ^{222}Rn fluxes ($12 \cdot 10^{-3}$–$25 \cdot 10^{-3}$ Bq m^{-2} s^{-1}) on loamy and clayey soils (M1 and M2).

In relation with the temporal variation of the ^{222}Rn release for each soil, the ^{222}Rn release from the clayey soil shows higher variations than the sandy soils (see Table 4). Consequently, the ^{222}Rn release from clayey soils should present seasonal variations more significantly than sandy soils. This behavior is corroborated in this because the ^{222}Rn release from clayey soils exhibits better correlation with the characteristic paraeters of soil and meteorological parameters than the ^{222}Rn release from sandy soils. This behavior is shown in the following paragraph.

4.2 Factors affecting values of ^{222}Rn release

In this study, we have excluded the ^{222}Rn release from soil M3 because the number of measurements were scarce.

Table 4. Fitting results of ^{222}Rn release (J_{Rn}) with soil humidity (H) by linear regression analysis: $J_{Rn} = mH + b$

Soil type	Slope (m)	Intercept (b)	r	p (%)
M1	$-(3.8 \pm 0.5)\cdot 10^{-4}$	$(15.2 \pm 0.5)\cdot 10^{-3}$	-0.9878	>99.8
M2	$-(2.7 \pm 0.3)\cdot 10^{-4}$	$(26.8 \pm 0.6)\cdot 10^{-3}$	-0.9756	>99.9
M4	$-(2.3 \pm 1.1)\cdot 10^{-4}$	$(11.5 \pm 1.3)\cdot 10^{-3}$	-0.8245	>80

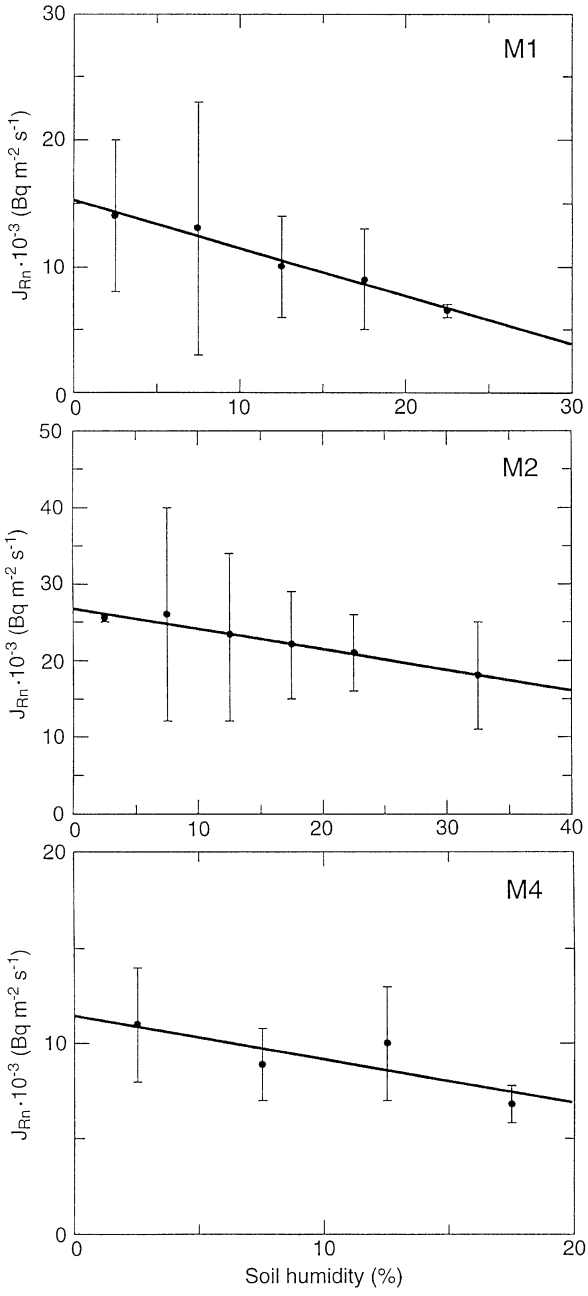

Fig. 2. Variations in ^{222}Rn release with respect to soil humidity

4.2.1 ^{222}Rn release from soils and soil humidity
In M1 and M2 we have obtained an inverse correlation between ^{222}Rn release and soil humidity; in M4, this behavior is analogous although the degree of confidence is smaller.

Figure 2 shows the correlations between the ^{222}Rn release and soil humidity on soils M1, M2 and M4. Table 4 exhibits the coefficients of equation of regression (m and b), the correlation coefficient (r) and the degree of confidence (p) for the soils mentioned.

Soil humidity appears as the decisive factor in release values. Generally, the presence of water in soil produces a reduction of soil porosity and consequently a diminution in gas diffusion rates, in particular, in the top decimeter of soil. This retards ^{222}Rn release and increases the concentration of ^{222}Rn in the top layer near the interphase soil-atmosphere. In soil with a high degree of permeability, the presence of water in soil is less important than for soils with low permeability values. The soils M1 and M2 have, respectively, slight and small permeability, while M4 is moderately permeable. For this reason, the correlation between ^{222}Rn release and soil humidity is better for M1 and M2 than M4. This behavior is analogous to those reported by other investigators, Cox *et al.* (1970), Megumi and Mamuro (1972), Martín Salas *et al.* (1979) and Quindós (1980).

4.2.2 ^{222}Ra release and soil thermic gradient
We have found a correlation between ^{222}Rn release and thermal gradient for soil temperature between 15 and 80 cm deep for soils M1 and M2. The results of this correlation are shown in Fig. 3 and Table 5.

The direct correlation between ^{222}Rn release and the soil thermal gradient has originated because an increasing surface soil temperature produces an enhanced surface release. Moreover, the relation between ^{222}Rn releases and soil thermal gradient is consequence of soil humidity. When the temperature of the top soil is high, the soil humidity is low and consequently ^{222}Rn release would be greater than when the temperature of the top soil is low, a situation associated with negative thermal gradients linked generally to high soil humidity. This behavior is analogous to those reported by Martín Salas *et al.* (1979) and Fernández (1981).

4.2.3 ^{222}Rn release and atmospheric pressure
The existence of a significant atmospheric pressure effect is not surprising because many experiments have shown this relationship. Moreover, mathematical models predict such an effect (Clements and Wilkening, 1974; Edwards and Bates, 1980). Figure 4 shows ^{222}Rn release measured at 1500 (UT) in each case as a function of the barometric pressure averaged over 12 h before the sampling was completed for M1 and M2 soils. Table 6 shows the characteristic parameters of adjustments.

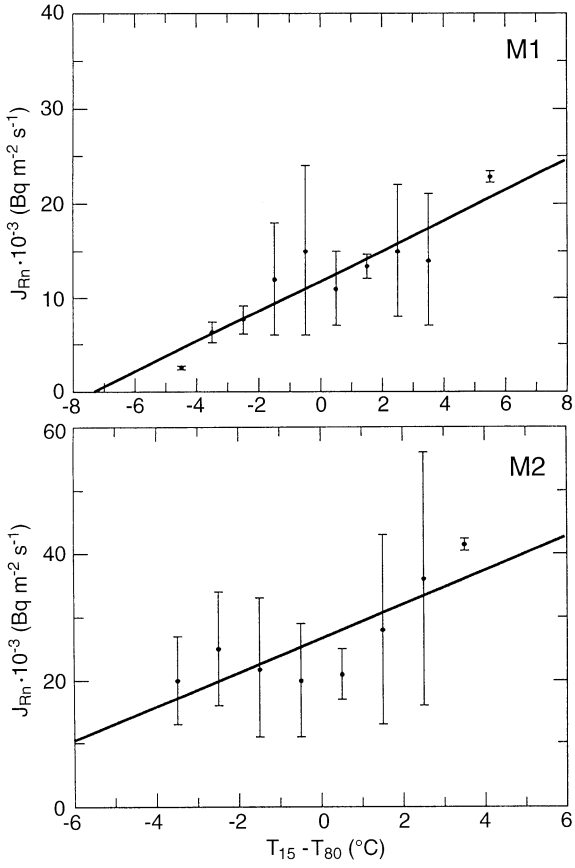

Fig. 3. Relation between ^{222}Rn release and thermal soil gradient for soils M1 and M2

Table 5. Fitting results of ^{222}Rn release (J_{Rn}) with soil thermal gradient (ΔT) by linear regression analysis: $J_{Rn} = m\Delta T + b$

Soil type	Slope (m)	Intercept (b)	r	p (%)
M1	$(1.6 \pm 0.3) \cdot 10^{-3}$	$(11.8 \pm 0.8) \cdot 10^{-3}$	0.9125	> 99.9
M2	$(2.7 \pm 0.8) \cdot 10^{-3}$	$(26.7 \pm 1.8) \cdot 10^{-3}$	0.8125	> 98.0

4.2.4 ^{222}Rn release and wind speed

Model calculations for wind to be reported in detail elsewhere indicates that its effect is to increase release. Enhancement of release due to wind has been previously reported (Kovach, 1946; Martín Salas *et al.*, 1979; Dueñas and Fernández, 1983). In this study we have found a direct correlation between ^{222}Rn release and the wind speed averaged over 2 h before sampling only for soil M2. Moreover, this correlation has been obtained by fixing the soil humidity at values greater than or equal to 10%. We suppose that the correlation obtained is coherent becausse the increase of wind speed is linked to turbulent mixing that increases the ^{222}Rn gradient between soil and air and this enhances ^{222}Rn release. Figure 5 shows the correlation between ^{222}Rn release and wind speed ($\bar{v}$) for soil M2. Table 7 presents the coefficients of equation of regression (m and b), the correlation coefficient (r) and the degree of confidence (p) for soil M2.

We conclude that soil parameters and variable meteorological effects influence ^{222}Rn release from soils M1 and M2 more than from M4. ^{222}Rn release from soils M1 and M2 presents oscillations higher than from M4, consequently, the ^{222}Rn release from soils M1 and M2 would be more affected by the parameters considered than M4 soil.

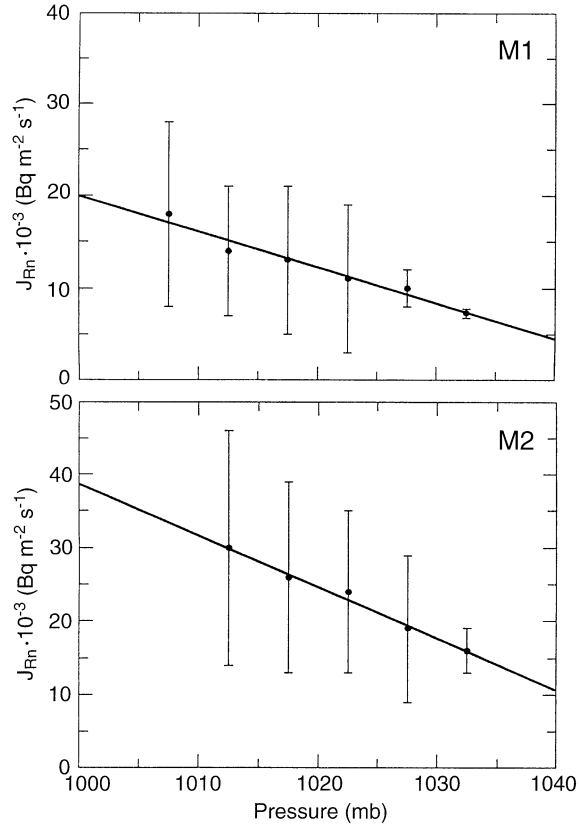

Fig. 4. ^{222}Rn release as a function of atmospheric pressure for soils M1 and M2

Table 6. Fitting results of ^{222}Rn release (J_{Rn}) with barometric pressure ($\bar{p}$) by linear regression analysis: $J_{Rn} = m\bar{p} + b$

Soil type	Slope (m)	Intercept (b)	r	p (%)
M1	$-(3.9 \pm 0.4) \cdot 10^{-4}$	0.41 ± 0.04	-0.9797	> 99.9
M2	$-(7.0 \pm 0.4) \cdot 10^{-4}$	0.74 ± 0.05	-0.9939	> 99.9

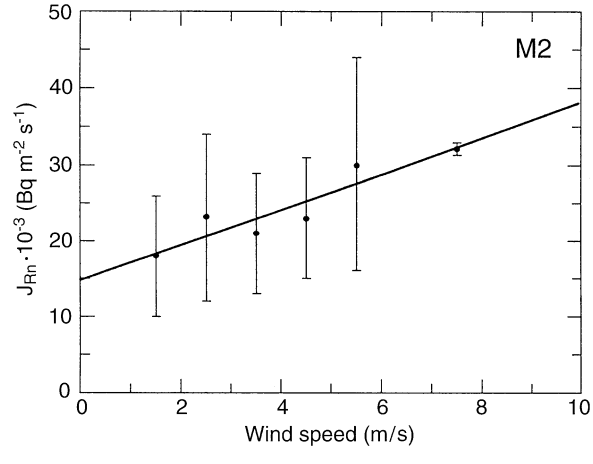

Fig. 5. ^{222}Rn release and wind speed for soil M2

Table 7. Fitting results of ^{222}Rn exhalation (J_{Rn}) with wind speed (v) by linear regression analysis: $J_{Rn} = mv + b$

Soil type	Slope (m)	Intercept (b)	r	p (%)
M2	$(2.3 \pm 0.5) \cdot 10^{-3}$	$(14.8 \pm 2.2) \cdot 10^{-3}$	0.9277	> 99.0

In particular, for sandy soil Fernández *et al.* (1983), Schery *et al.* (1984) and Dörr and Münnich (1990) have not found a correlation between ^{222}Rn release and meteorological parameters; Fernández *et al.* (1983) observed the influence of soil humidity on ^{222}Rn release from a sandy soil at a sampling point with a very wet climate; Schery *et al.* (1984), for a sandy soil with semi-arid climate, found correlation between ^{222}Rn release and barometric pressure; finally, Dörr and Münnich (1990) did not find a correlation between ^{222}Rn release and parameters, such as meteorological variations and soil characteristics in a wet climate.

5 The concentration profiles of ^{222}Rn in the soil air

In soil M1, measurements of ^{222}Rn in soil air concentrations were made by inserting tubes into the soil at 18, 47, 75, 150 and 222 cm in depth; in M2, at 10, 15, 30, 45 and 60 cm in depth; in M3, at 5, 10, 15, 30, 45, 60, 85 and 105 cm in depth and in M4 at 5, 10, 15, 30, 45, 60, 120, 180, 240 and 325 cm in depth.

The mean concentration profiles of ^{222}Rn concentration in the study soils are given in Fig. 6. ^{222}Rn concentration profiles increases with depth to a constant value, $C_{\infty, Rn}$, which varies according to the soil types. The solid curves indicate the ^{222}Rn concentration profiles fitted to an exponential function by least squares method, Eq. (4).

Table 8 shows the characteristic parameters of the adjustments carried out, C_∞ and $\bar{z}$ for ^{222}Rn. The average relaxation depth is in good agreement with the theoretical value of 100 cm for ^{222}Rn (Nazaroff *et al.*, 1988). Also, the number of profiles achieved (N), the chi squared (χ^2) and the degree of confidence (p) results are given in Table 8.

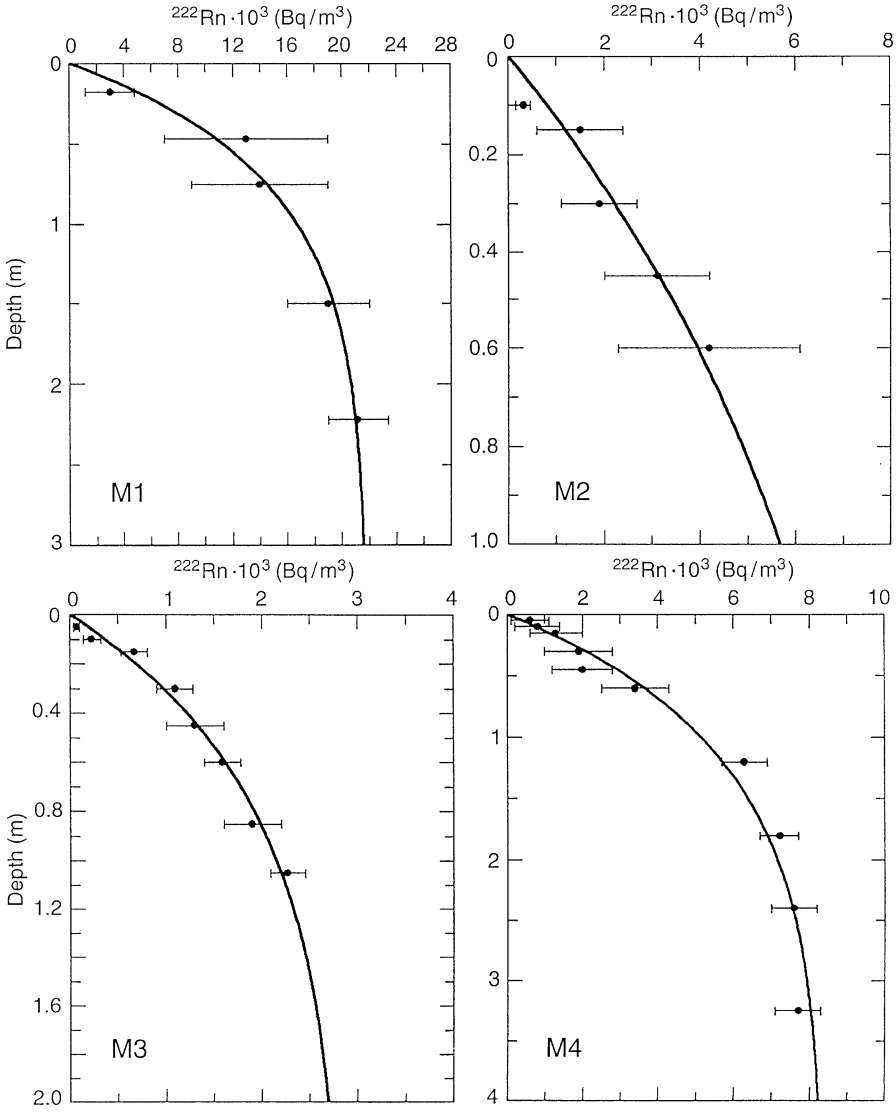

Fig. 6. Profiles of Rn concentration in the soil air for soils studied

Table 8. Fitting results of ^{222}Rn concentration profiles in air soil

Location	N	C_∞ (Bq/m^3)	$\bar{z}$ (cm)	χ^2	p (%)
M1	63	21840 ± 160	68.4 ± 1.4	1.44	>99.5
M2	45	11800 ± 600	152 ± 14	9.84	97.5
M3	20	2880 ± 100	72 ± 4	20.34	99
M4	40	8430 ± 70	105.6 ± 2.2	3.36	>99.5

Table 9. Porosity and diffusion coefficient for each soil

Soil type	M1	M2	M3	M4
ε (%)	25.9 ± 0.3	42.9 ± 0.1	36.4 ± 0.4	43.4 ± 0.3
$D_{Rn} \cdot 10^{-3}$ (cm^2/s)	2.5 ± 0.1	21 ± 4	4.0 ± 0.4	10.2 ± 0.4

The results of adjustments are very acceptable because the majority exhibit a degree of confidence greater than or equal to 99%; this behavior confirms the supposed hypothesis that the transport in the unsaturated zone is really due to molecular diffusion.

5.1 Diffusion coefficient

We evaluated the average diffusion coefficient in each soil from Eq. (5), using measured porosity (ε) and the calculated value of relaxation depth ($\bar{z}$). Table 9 shows the porosity and the diffusion coefficient for each soil studied.

Using Table 9, we conclude that the average diffusion coefficient for soils M2 and M4 was $15.6 \cdot 10^{-3}$ cm^2/s and $3.25 \cdot 10^{-3}$ cm^2/s for soils M1 and M3. Considering that the molecular diffusion coefficient of ^{222}Rn in the air may be taken as 0.1 cm^2/s and in the water as 10^{-5} cm^2/s (Tanner, 1964), we thought that the obtained values for diffusion coefficient should be related to the average humidity of soil during the measurement period. Evaluating the average humidity for each soil, we have not found any important differences between the humidities of the different soils to explain the wide range of values that we found on soils studied for diffusion coefficients.

The soils gas sampling at the greatest depth was different for each soil, when we analyzed the diffusion coefficient for each soil together with the greater depth, their location and soil properties. The M2 site provided a clayey soil, covered by old trees (Ficus and Eucalytus). The soil gas sampling at depths of 10, 15, 30, 45 and 60 cm from installations permanently situated on tree roots, which joined with possible internal retractions produced by their clayey nature, may be the factors that help increase the diffusion of ^{222}Rn between 60 cm depth and the surface. The M2 site presents the highest value of diffusion coefficient. The M1 site presents a very heterogeneous structure in relation to their composition and grain size with clay on the surface and a mixture of sand and gravel in depth. The soil gas sampling at depths of 18, 47, 75, 150 and 122 cm are from permanent installations. The M1 site exhibits the smallest values of diffusion coefficients. The M3 sandy soil also presents a small diffusion value of coefficient; this value may be attributed to their typical location and the rains that occured closely the sampling time. The soil M4 presents a diffusion coefficient that is with in the range of sandy soils with small humidity soil (Dueñas and Fernández, 1987).

Table 10. Emanation fraction of ^{222}Rn from soils studied

Soil type	M1	M2	M3	M4
f (%)	10.7 ± 0.7	6.8 ± 0.6	2.3 ± 0.2	9.6 ± 0.6

5.2 Emanation fraction of ^{222}Rn from sampling soils

We have calculated from Eq. (6) the emanation fraction or emanation coefficient of ^{222}Rn using measured radium content, the bulk density, the porosity and the calculated value of $C_{\infty, Rn}$ for the four study soils. These results, summarized in Table 10, indicate a range of 2.3 to 10.7%.

The average value obtained for the emanation fraction for the soils sampled was 7%. According to Rama and Moore (1984), the fraction emanation is between 1–10% depending on the effective soil particle surface and thus, on grain size distribution. The M3 soil presents the smallest emanation fraction and precisely this soil has also the smallest ^{222}Rn release. Several factors have been demonstrated in several studies to have a large impact on the emanation coefficient, such as the moisture content and the grain size distribution (Nazaroff *et al.*, 1988). When estimating the different parameters used to evaluate the emanation fraction of ^{222}Rn, Eq. (6), the main differences are of the values of C_∞ and the soils that exhibit the greatest (M4) and smallest value (M3) of the radioactive equilibrium concentration are precisely those which have the highest and lowest emanation fractions respectively.

5.3 Comparison between ^{222}Rn obtained fluxes for direct and indirect method

The ^{222}Rn fluxes have been evaluated by two methods. A direct method, accumulation, before exposed, and an indirect method from Eq. (7). Table 11 shows the average ^{222}Rn fluxes by direct and indirect methods for all investigated sites. In all soils sampled, the flux evaluated by direct method is higher than the flux obtained by indirect method. These results are analogous to those reported by other papers (Kraner *et al.*, 1964; Schroeder *et al.*, 1965; Schery *et al.*, 1984).

The greatest difference between direct and indirect flux is exhibited by M3 soil; this difference may be attributed to its locational peculierities already mentioned. The flux evaluated by indirect method has been obtained by supposing that the transport of ^{222}Rn in the soil air is controlled by molecular diffusion. Thus, the results obtained may indicate that there may be other mechanisms of transport operating in addition to diffusion such as the convective processes or some fissures that augment the flux obtained by direct method.

Table 11. The average ^{222}Rn fluxes by indirect and direct method

Soil type	$\bar{J}_{Rn}$ (Bq m^{-2} s^{-1}) Direct method	$\bar{J}_{Rn}$ (Bq m^{-2} s^{-1}) Indirect method
M1	$(12 \pm 8)\cdot 10^{-3}$	$(8.1 \pm 0.3)\cdot 10^{-3}$
M2	$(25 \pm 13)\cdot 10^{-3}$	$(16.1 \pm 2.3)\cdot 10^{-3}$
M3	$(3.0 \pm 1.6)\cdot 10^{-3}$	$(1.60 \pm 0.18)\cdot 10^{-3}$
M4	$(10 \pm 4)\cdot 10^{-3}$	$(8.0 \pm 0.3)\cdot 10^{-3}$

In soils M1 and M2 the quotient between direct and indirect flux is higher than the same quotient for soil M4. In M1 and M2 we have found a good linear correlation between the direct flux of ^{222}Rn and the thermal gradient of soil between 15 and 80 cm (already exposed), while for M4 we have not found any correlation for mentioned parameters.

6 Conclusions

The data obtained characterize the flux from the surface of the most widespread generic soil of Málaga. The factors that most affect the value of ^{222}Rn release were humidity and soil thermal gradient. Other factors that also affect the value of ^{222}Rn release are barometric pressure and wind speed. The directly measured ^{222}Rn fluxes at investigated sites are higher than ^{222}Rn fluxes derived by the indirect method.

Acknowledgements. Topical Editor L. Eymard thanks Y. Balkanski and J. Fontan for their help in evaluating this paper.

References

Carretero, J., Flujos y perfiles de Rn, CO_2 y CH_4 en el suelo, Doctoral Thesis, University of Granada, Spain, Granada 1994.

Clements, W. E., and M. H. Wilkening, Atmospheric pressure effects on ^{222}Rn transport across the earth-air interface, *J. Geophys. Res.*, **79,** 5025–5029, 1974.

Cox, W. M., R. L. Blanchard, and B. Kahn, Relation of Radon concentration in the atmosphere to total moisture detection in soil and atmospheric thermal stability, in *Radionuclides in the environment*, Avances in Chemistry Series, **93,** 436–446, 1970.

Dörr, H., and K. O. Münnich, ^{222}Rn flux and soil air concentration profiles in West Germany. Soil ^{222}Rn as tracer for gas transport in the unsaturated soil zone, *Tellus*, **42B,** 20–28, 1990.

Druilhet, A., and J. Fontan, Determination des coefficients de diffusion verticle enture 0 et 100 m, *Boundary Layer Meteorol.*, **3,** 476–495, 1972.

Dueñas, C., and M. C. Fernández, Medida de la exhalación en Rn del suelo e influencia sobre la misma de ciertas variables, *An. Fis.*, **B, 79,** 10–17, 1983.

Dueñas, C., and M. C. Fernández, Dependence of Radon-222 flux on concentrations of soil gas and air and an analysis of the effects produced by several atmospheric variables, *Ann. Geophysicae*, **5B6,** 533–539, 1987.

Dueñas, C., and M. C. Fernandez, Temporal variations in soil gas radon: any possible relation to earthquakes?, *Tectonophysics*, **152,** 134–145, 1988.

Edwards, J. C., and R. C. Bates, Theoretical evaluation of Radon emanation under a variety of conditions, *Health Phys.*, **39,** 263–274, 1980.

Fernández, M. C., Medida de la exhalación en Radón de suelo y del mar. Influencia sobre la misma de diversos factors, Doctoral Thesis, University of Málaga, Spain, 1981.

Fernández, P., L. S. Quindós, J. Soto, E. Villar, and D. Guedalia, A new method for measuring Radon exhalation, *J. Geophys. Res.*, **88,** C2, 1519–1524, 1983.

Graustein, W. C., and K. K. Turkekian, Radon fluxes from soils to the atmosphere measured by ^{210}Pb–^{226}Ra disequilibrium in soils, *Geophys. Res. Lett.*, **17,** 841–844, 1990.

Guedalia, D. A. A. Ntsila, A. Druilhet, and J. Fontan, Monitoring of the atmospheric stability above an urban and suburban site using sodar and radon measurements, *J. Appl. Meteorol.*, **19,** 839–845, 1980.

Hsu, S. A., R. E. Larson, and D. J. Bressan, Diurnal variation of radon and mixing heights along a coast: a case study, *J. Geophys. Res.*, **85,** 4107–4112, 1980.

Israël, H., Radioactivity of the atmosphere, T. F. Malone (ed) Compendium of Meteorology, *Am. Meteorol. Boston*, 155–161, 1951.

Jiménez, J. A., and J. L. de Justo, Propiedades de los suelos y de las rocas, Ed. Rueda, Madrid, 1975.

Karol, I. L., Radioisotopes and global transport in the atmosphere, 44–217, Israel Program for Scientific Translations, Jerusalem, 1974.

King, C. Y., Radon emanation on San Andreas, *Nature*, **271,** 516–519, 1978.

Kovach, E. M., Diurnal variations of the radon content of soil-gas, *Terr. Magnet, Atmos. Electr.*, **51,** 45–55, 1946.

Kraner, H. W., G. L. Schroeder, and R. D. Evans, Measurements of the effects of atmospheric variables on radon-222 flux and soils gas concentrations, in *The natural radiation environment*, University of Chicago Press, 191–215, 1964.

Lambert, G., G. Polian, J. Sanak, B. Ardouin, A. Buissen, A. Jègen, and J. C. Le Roulley, Cycle du Radon et ses descendants: application à l'étude des èchanges troposphère-stratosphère, *Ann. Géophys.* **38,** 497–531, 1982.

Larson, R., and P. Bressan, Radon-222 as an indicator of continental air masses and air mass boundaries over ocean areas, in *The natural radiation environment*, vol. 3. (Ed.) T. Ysell, W. Lodwer, National Technical Information Service, Springfield, Virginia, 1980.

Lucas, H., Improved low-level alpha scintillation counter for radon, *Rev. Sci. Instrum.*, **28,** 1957.

Martín Salas, A., M. Pardos, and J. Casanova, Medida de la exhalación del Radón ^{222}Rn y estudio de la influencia sobre la misma de ciertas condiciones meteorológicas y del suelo, *An. Fis.*, **75**(3) 135–139, 1979.

Megumi, K., and T. Mamuro, A method for measuring radon and thoron exhalations from the ground, *J. Geophys. Res.*, **74**(N17) 3052–3056, 1972.

Mochizuki, S., and T. Sekikawa, Radon-222 axhalation rate and its variation in soil air, in *Natural radiation environment III*, conf. 780422, U.S.D.E. 1, 105–116, 1978.

Nazaroff, W. W., B. A. Moed, and R. G. Sextro, Soil as source of indoor radon: generation, migration and entry, in *Radon and its decay products in indoor air*. (Ed.) W. W. Nazaroff, A. V. Nero Jr, John Wiley, New York, 57–112, 1988.

Nero, A. V., and W. W. Nazaroff, Characterizing the source of Radon indoors, *Radiat. Prot. Dosim.*, **7,** 23–29, 1983.

Pearson, J. E., and E. Jones, Emanation of radon-222 from soils and its use as a tracer, *J. Geophys. Res.*, **70,** 5279–5290, 1965.

Polian, G. P. Monfray, B. Ardouin, and G. Lambert, Modèlisation du transport à longue distance des masses d'air marquées par le Radon-22;2, in *Actes du Colloque sur la recherche française dans les terres australes*, Publication Terrotoire des TAAF, Paris, 417–428, 1989.

Prospero, J. M., and T. N. Carlson, Radon 222 in the North Atlantic trade winds; its relationship to dust transport from Africa, *Science*, **167,** 974–977, 1970.

Quindós, L. S., Desarrollo y puesta a punto de un nuevo método para la medida de la exhalación de Radón, Doctoral Thesis, University of Santander, Spain, 1980.

Quindós, L. S., P. L. Fernandez, J. Soto, and G. Newton, A modified Lucas cell for leakage measurement from encapsulated radium sources, *Appl. Radiat. Isot.*, **42,** 1108–1112, 1991.

Rama, and W. S. Moore, Mechanism of transport of U-Th series radioisotopes from solids into ground water, *Geochim. Cosmochim. Acta*, **48,** 395–399, 1984.

Schery, S. D., D. H. Gaeddert, and M. H. Wilkening, Factors affecting exhalation of radon from a gravelly sandy loam, *J. Geophys. Res.*, **89,** 7299–7309, 1984.

Schroeder, G. L., H. W. Kraner, and R. D. Evans, Diffusion of radon in several naturally occurring soil types, *J. Geophys. Res.*, **70,** 471–478, 1965.

Servant, J., Le Radon et ses derivés à vie courte dans l'atmosphère, Thèse de Doctorat ès Sciences, Université de Paris, France, 1964.

Sisigina, T. I., Radon emanation from the surface of some types of soils of the European part of the USSR and Kazakhstan, in *Radioactive isotopes in the atmosphere and their use in meteorology*, (Ed.) I. L. Harol *et al.*, Washington D.C., 29–33, 1967.

Tanner, A. B., Radon migration in the ground: a review, in *The natural radiation environment*, 161–190, University of Chicago Press, Chicago, 1964.

Turekian, K. K., Y. Nozaki, and L. K. Benninger, Geochemistry of atmospheric radon and radon products, *Annu. Rev. Earth Planet. Sci.*, **5,** 227–255, 1977.

Wilkening, M. H., W. E. Clements, and D. Stanley, Radon-222 flux measurements in widely separated regions, in *The natural radiation environment II*, (Ed.) John A. S. Adams, Rice University Houston, Texas, 717–730, 1972.

Wilkniss, P. E., R. E. Larson, P. J. Bressan, and J. Steranka, Atmospheric radon and continental dust near the automatic and their correlation with air mass trajectories, *J. Appl. Meteorol.*, **13,** 512–520, 1974.