

# Radon migration in the soils of the Irno Valley (Southern Italy) inferred from radioactive disequilibrium

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## Abstract

Radon migration along vertical profiles in the soils of Irno River alluvial Valley (Southern Italy) was studied using radioactive disequilibrium between  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$ . Fractional Radon loss, migration length, diffusion and emanation coefficient and Radon flux density were determined. Our results are in agreement with a migration model by simple diffusion. The migration parameters are within typical values, except the Radon flux density, which is about one order of magnitude higher than the values reported in literature. The values of fractional Radon loss are sensitive to changes in the physical properties of the soil.

**Key words** Radon – disequilibrium – Radon migration

## 1. Introduction

Radon is the main component of natural radioactivity. Its most abundant isotopes are  $^{222}\text{Rn}$ , which is produced by the decay of  $^{238}\text{U}$ , and  $^{220}\text{Rn}$ , which is produced by  $^{232}\text{Th}$  daughters. Soils emit Radon, which migrates through the pores, reaches the solid Earth surface and passes to the atmosphere. Measurements of Radon concentration are of relevant interest for public health hazard. Radon also appears to be a tracer and a possible precursor of dynamical phenomena in the Earth, such as earthquakes and eruptions (see, for example, Gasparini and Veltri, 1987; Gasparini and Mantovani, 1978). All these applications require a knowledge of the mechanism of generation and transport of Radon in natural soils.

Theoretical and experimental studies on Radon have been reviewed by Tanner (1980)

and Nazaroff (1992), and further information is contained in the papers by Morawska and Phillips (1992), Schery *et al.* (1989) and Washington and Rose (1992).

Radon migration models assume generally a homogeneous, isotropic and isothermal soil. Under these conditions the key parameters ruling migration are the diffusion coefficient and the soil permeability which are strongly dependent on soil type and water content.

The best way to determine the parameters describing the mechanical transport of Radon would be to measure Radon concentrations at different depths. This would require a station equipped for a series of continuous measurements of Radon concentrations and meteorological parameters. A more convenient way is to use a method based on radioactive disequilibrium (Graustein and Turekian, 1990). This method utilizes a relatively long-life isotope of a solid element as a tracer for Radon presence. In fact the longest life isotope of Radon,  $^{222}\text{Rn}$ , ( $T_{1/2} = 3.82\text{ d}$ ) generates  $^{210}\text{Pb}$  ( $T_{1/2} = 22.3\text{ y}$ ) which is easily adsorbed on soil surfaces. Ac-

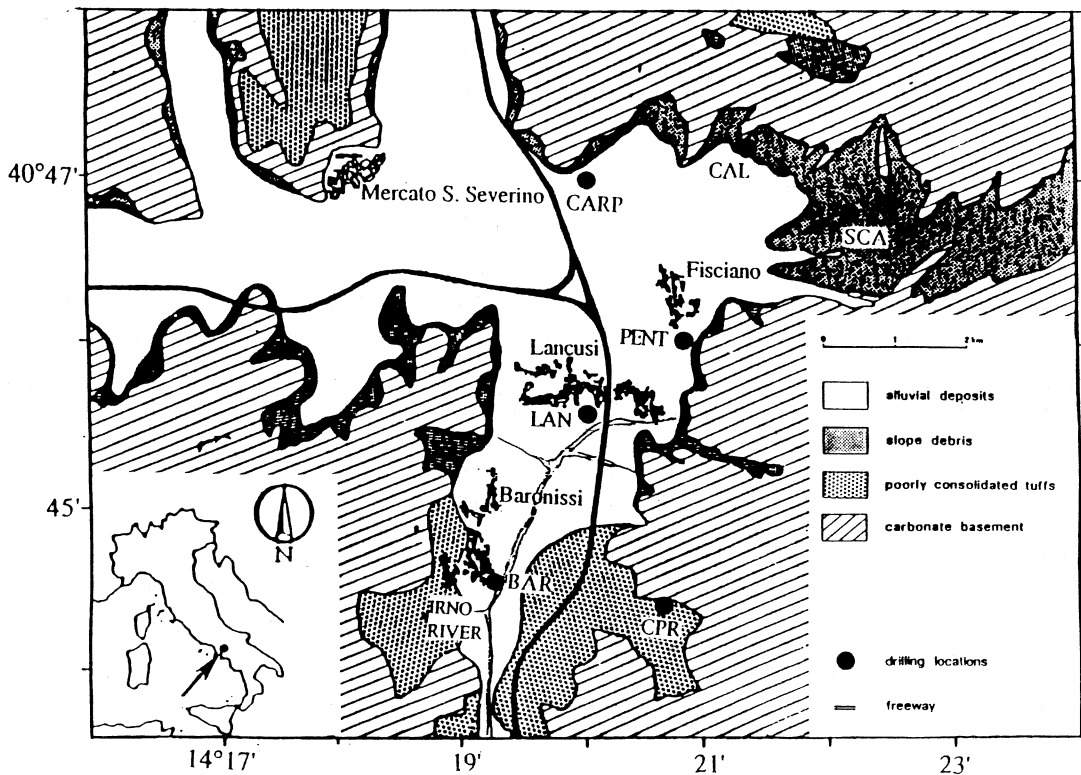
cordingly, its distribution with depth reflects the diffusional Radon loss, averaged over times long enough to smooth out local meteorological effects.

In the present paper the Radon migration characteristics in the soils covering a Quaternary alluvial valley surrounded by Mesozoic carbonate rocks are reported. They were determined measuring  $\gamma$ -activity due to  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$ . In a preliminary phase of the research the reproducibility of the results was checked in two areas by comparing the determinations from a set of eight holes drilled at very short distances from each other (Sabbarese *et al.*, 1993b).

The obtained data are used to calculate the significant migration parameters down to 4.4 m of depth.

## 2. Brief geological outlines of the Irno Valley

The Irno Valley (fig.1), is located north of Salerno (Southern Italy). It is filled mostly by river sediments interbedded by air fall pyroclastics coming from the neighbouring volcanoes of the neapolitan area. The valley is cut within a coastal mountain range formed by carbonatic rocks belonging to a Mesozoic marine platform. The sedimentary sequence filling the valley is covered by a thin layer of soil, which is underlain by unconsolidated air fall pyroclastics and alluvial deposits. This sequence is 20-40 m thick and lays on a lithic basement formed by volcanic tuff and, in the southern part of the valley, by carbonatic breccias. Limestones have been found at a depth of



**Fig. 1.** Lithological map of the Irno Valley. The circles and the corresponding codes indicate sites of measurements.

**Table I.** Values of soil parameters and Radon migration using fractional Radon loss in the superficial layers of the seven sites.

Site	$x$ (cm)	$\rho$ (g/cm <sup>3</sup> )	$\varepsilon$	[ <sup>226</sup> Ra] (Bq/kg)	$f$	1/g (cm)	$J$ (Bq/m <sup>2</sup> s)	$D$ (cm <sup>2</sup> /s)	$v$ (cm/s) $\times 10^{-5}$
CARP	0-80	1.25	0.69	290	0.71	73	0.20	0.0013	3.0
PENT	0-80	1.30	0.46	226	0.43	64	0.12	0.0011	3.6
LAN	0-90	1.20	0.37	196	0.70	77	0.11	0.0046	0.4
BAR	0-50	1.34	0.32	98	0.56	30	0.05	0.0024	2.3
CPR	0-120	1.36	0.64	121	0.68	107	0.10	0.0031	1.8
CAL	0-40	1.28	0.51	197	0.59	23	0.07	0.0045	0.9
SCA	0-300	1.41	0.47	205	0.55	140	0.11	0.0072	3.0

$x$  = Upper layer thickness;  $\rho$  = soil density;  $D$  = diffusion coefficient;  $v$  = velocity of fluid; 1/g = average migration length;  $J$  = Radon flux;  $\varepsilon$  = total porosity;  $f$  = emanation coefficient.

about 80-100 m in the inner part of the valley, and at greater depth in the Lancusi-Fisciano area, where a massive alluvial cone is present (Guzzetta, 1963).

Four of our Radon measurement sites (BAR, LAN, PENT, CARP) are located on alluvial sediments, two on the slope debris of a limestone hill (CAL, SCA) and one on a poorly consolidated volcanic tuff outcrop (CPR).

Total porosities, densities, water content and grain size of the soils were measured in the laboratory for each site. Densities are in the range 1200-1410 kg/m<sup>3</sup>, and total porosities range from 0.32 to 0.51 (table I). The correlation between density and porosity is not as good as it should be, because the soil is quite dishomogeneous having a variable volcanic component (Civetta *et al.*, 1970; Gialanella *et al.*, 1988).

### 3. Theoretical model of Radon migration in soils

Radon is assumed to migrate within a homogeneous, isotropic and porous medium, with semi-infinite length (as discussed by Clements and Wilkening, 1974), by diffusion described

by Fick's law and by advective transport described by Darcy's law. The assumed homogeneity for the soil is not in disagreement with the above mentioned experimental inhomogeneities. As a matter of fact such inhomogeneities have a scale smaller than that considered in our study.

The variation with depth of the Radon content within the soil's pores is described by the steady-state solution of the following differential equation:

$$\frac{\partial C(x, t)}{\partial t} = \frac{D}{\varepsilon} \frac{\partial^2 C(x, t)}{\partial x^2} + \frac{1}{\varepsilon} \frac{\partial [v(x, t) \cdot C(x, t)]}{\partial x} - \lambda C(x, t) + \phi(x) \quad (3.1)$$

where  $C$  is the Radon specific activity (Bq/m<sup>3</sup>),  $D$  is the Radon diffusion coefficient in the soil (m<sup>2</sup>/s),  $\varepsilon$  is the total porosity,  $v$  is the velocity of fluid (m/s),  $\lambda$  is the decay constant of <sup>222</sup>Rn (s<sup>-1</sup>),  $\phi$  is <sup>222</sup>Rn input rate in the pores (Bq/m<sup>3</sup>s). The  $x$ -axis is chosen perpendicular to the Earth surface with origin on the air-soil interface and the positive direction downward.

The boundary conditions are:

$$C(0, t) = 0$$

$$C(\infty, t) = \phi/\lambda.$$

The steady-state solution of this equation, with constant  $D$ ,  $v$  and  $\phi$  is the following:

$$C(x) = (\phi/\lambda) \{1 - e^{-gx}\} \quad (3.2)$$

where

$$g = (v/2D) + \sqrt{(v/2D)^2 + \epsilon\lambda/D}, \quad (3.3)$$

$g$  is the inverse of the mean length of Radon migration.

#### 4. The $^{210}\text{Pb}$ - $^{226}\text{Ra}$ disequilibrium method

$^{222}\text{Rn}$  is generated from  $^{226}\text{Ra}$  ( $T_{1/2} = 1600\text{y}$ ) by  $\alpha$  decay. The chain continues with four elements of short half-life which ultimately decay to  $^{210}\text{Pb}$  ( $T_{1/2} = 22.3\text{y}$ ). This lead isotope can be expected to persist in measurable amounts for about 40 years in the sites where Radon decayed.

The generated  $^{222}\text{Rn}$  divides into two fractions; one moving through the rock pores and the other being trapped in the soil grains. The former is equal to the  $^{226}\text{Ra}$  activity multiplied by the emanation coefficient of soil ( $f$ ):

$$[^{222}\text{Rn}] = f[^{226}\text{Ra}] \quad (4.1)$$

where square brackets denote specific activity.

The total  $^{210}\text{Pb}$  concentration in the soil is also divided into two fractions, one being generated by the Radon trapped in the grains and the other by the Radon moving through the soil pores. The latter is adsorbed on the solid surfaces of the soil. Hence, combining eqs. (3.2) and (4.1):

$$\begin{aligned} [^{210}\text{Pb}] &= [^{222}\text{Rn}]_{\text{grains}} + [^{222}\text{Rn}]_{\text{pores}} = \\ &= [^{226}\text{Ra}] \cdot (1 - f \cdot e^{-gx}). \end{aligned}$$

The fractional deficiency of  $[^{210}\text{Pb}]$ , defined as:

$$[Pb_d] = \frac{[^{226}\text{Ra}] - [^{210}\text{Pb}]}{[^{226}\text{Ra}]} = f \cdot e^{-gx} \quad (4.2)$$

is a measure of the fractional Radon loss.

$[^{210}\text{Pb}]$  values in depth profiles are generally less than  $[^{226}\text{Ra}]$  values indicating the escape of a consistent fraction of Radon, except in the very shallow layer. Here the fallout of  $^{210}\text{Pb}$  from the atmosphere produces an excess over  $^{226}\text{Ra}$ , hiding the activity related to natural migration from underground. Graustein and Turekian (1990) showed the contribution of fall out  $^{210}\text{Pb}$  in soils can be accounted for using  $^{137}\text{Cs}$ , which is still in the atmosphere after nuclear weapons tests and nuclear power plants disasters (Gialanella *et al.*, 1988).  $^{137}\text{Cs}$  is in fact deposited from the atmosphere in a similar fashion to  $^{210}\text{Pb}$ . Graustein and Turekian (1990) use the assumption that disturbances from atmospheric  $^{210}\text{Pb}$  are relevant when  $[^{137}\text{Cs}]/[^{226}\text{Ra}] > 0.05$ . This usually happens in the shallowest 30 cm of soil. It is reasonable to assume that atmospheric conditions and water percolation distribute the two radionuclides in a similar way. For this reason, we correct the  $^{210}\text{Pb}$  data using the  $^{137}\text{Cs}$  data trend, which generally is well fitted by a decreasing exponential curve.

The best-fitting (made calculating the minimum of error function with conjugated gradient method) of the data measured along a vertical profile with eq. (4.2) allows the determination of the parameters ruling the generation and transport of Radon ( $f$ ,  $D$ ,  $v$  and  $g$ ).

The flux  $J$  (expressed in  $\text{Bq/m}^2\text{s}$ ) of Radon coming out of a soil of thickness  $x$ , can be calculated knowing the fractional Radon loss, Radium content and density of the soil from the following equation:

$$\int_0^x \frac{\partial J}{\partial x} = \frac{f \cdot \lambda \cdot \rho \cdot [^{226}\text{Ra}]}{g} (e^{-gx} - 1). \quad (4.3)$$

#### 5. Experimental techniques

Holes were located in sites which are not covered by agricultural soils, which have not

undergone significant erosion or sedimentation, and where no human activity was present so that the distributions of [ $^{210}\text{Pb}$ ], [ $^{226}\text{Ra}$ ] and [ $^{137}\text{Cs}$ ] reflect only the natural migration process. In this way, [ $^{210}\text{Pb}$ ] equals the long-term average [ $^{222}\text{Rn}$ ] trapped in the soil. 334 soil cores were collected in the seven sites of the Irno Valley indicated in fig. 1.

At CARP and PENT sites cores were obtained, respectively, from eight and seven 80 cm deep holes distributed over a 30 m<sup>2</sup> area at each site. Cores at LAN were obtained from two 120 cm deep holes, at BAR at CPR from two 220 cm deep holes at each site, at CAL from two 290 cm deep holes and at SCA from one 440 deep hole.

Soil cores were divided into several 10 cm sections (only at SCA sections were 20 cm long). These sections were dried up in the laboratory and sieved to remove rock fragments and to obtain the desired granulometry. These operations do not change the measured quantities by  $\gamma$  spectrometry. Finally, they were sealed in a 750 cm<sup>3</sup> Marinelli beaker for more than 20 days (about 5 times the mean-life of  $^{222}\text{Rn}$ ) to allow  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  to grow into equilibrium with  $^{226}\text{Ra}$ . We measured [ $^{210}\text{Pb}$ ], [ $^{226}\text{Ra}$ ] and [ $^{137}\text{Cs}$ ] by gamma-spectrometry, using an intrinsic germanium detector with a resolution of 1.9 keV at 1.33 MeV, properly shielded from background radiation. The gamma-ray counting-time of each sample was at least 12 h to obtain good precision of measurement. [ $^{210}\text{Pb}$ ] and [ $^{137}\text{Cs}$ ] were determined from their gamma-peaks at 46.5 and 661.6 keV, respectively. [ $^{226}\text{Ra}$ ] was obtained from the average value of the most intense gamma-lines of  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  at 352 and 609 keV, respectively.

The detection efficiency was determined using a standard soil source, manufactured by the Environmental Measurements Laboratory of CRE-Casaccia (ENEA). The correction for attenuation by self-absorption of gamma-rays was made by the method of Cutshall *et al.* (1983).

The statistical counting errors of different nuclides, for a concentration of 100 Bq/kg and 12 h of counting, were 2%, 0.7%, 0.9% and 0.6% for  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{137}\text{Cs}$ , respectively.

Emanation coefficients were determined using an electrostatic collector coupled to a surface barrier silicon detector (Sabbarese *et al.*, 1993a). This monitor directly detects the particles emitted by  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  daughters. For emanation measurements in the laboratory, the chamber was positioned on a thin layer of soil sample. At equilibrium the Radon concentration in the chamber equals the Radon in the soil pores. The emanation coefficient is calculated as the ratio between Radon in the chamber and Radium concentration of soil, determined by gamma spectrometry. Total porosity of soil was measured in the laboratory, on the same samples using a mercury porosimeter. Grain size distributions were measured using a series of sieves with ASTM classification. Density and water content of soils were measured using undisturbed samples taken *in situ* with small metallic cylinders.

## 6. Results and discussion

Variations of [ $^{226}\text{Ra}$ ], [ $^{210}\text{Pb}$ ] and [ $^{137}\text{Cs}$ ] with depth in the eight holes of CARP site are reported in fig. 2 together with the average fractional Radon loss and the best fitting curve. The pattern with depth is very similar for all the holes. The scatter of [ $^{226}\text{Ra}$ ] and [ $^{210}\text{Pb}$ ] can be ascribed to very local variation of the Radium rich volcanic component of the soil. [ $^{137}\text{Cs}$ ] is detectable down to 40 cm of depth, indicating that the fallout component and the corresponding correction of [ $^{210}\text{Pb}$ ] data are no longer important at deeper levels. This is consistent with [ $^{210}\text{Pb}$ ] being higher than [ $^{226}\text{Ra}$ ] down to 40 cm of depth and becoming lower than [ $^{226}\text{Ra}$ ] at deeper levels. [ $^{210}\text{Pb}$ ] in the shallowest 40 cm have been accordingly corrected for the fall-out component and the corrected specific activities have been used to calculate the fractional Radon loss.

Similar consistent patterns were also obtained for the seven holes at the PENT site (Sabbarese *et al.*, 1993b).

At all the other sites, except BAR, [ $^{137}\text{Cs}$ ] has the same pattern, decreasing exponentially with depth and being no longer detectable at 40 cm (an example is reported in fig. 3a). At

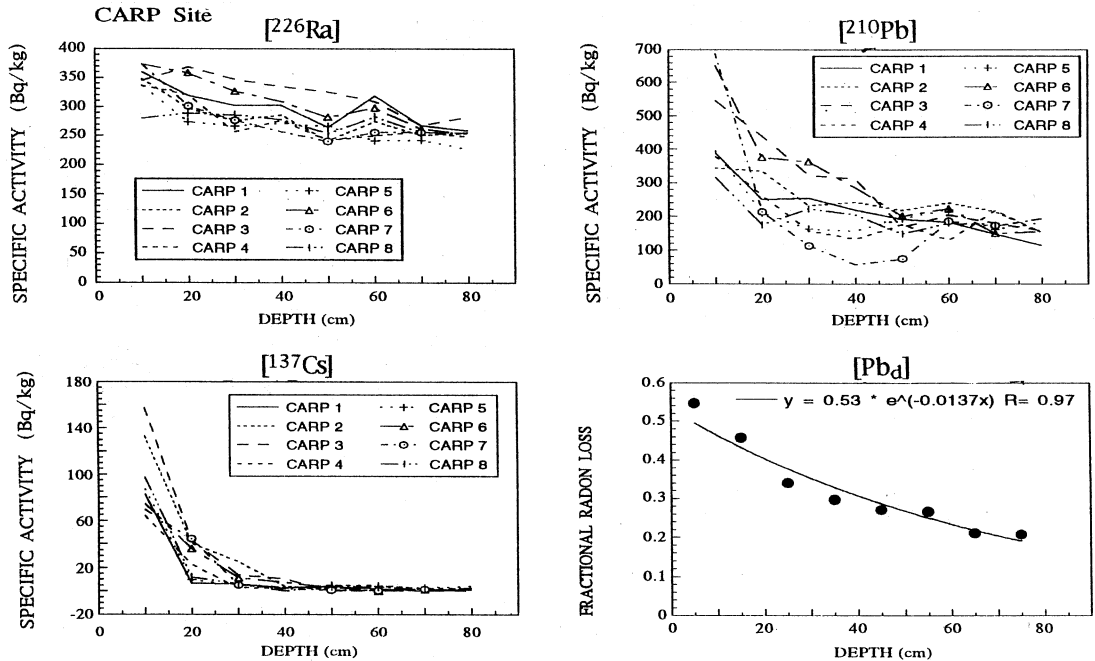


Fig. 2.  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$  and average fractional Radon loss variations with depth for the eight holes of the CARP site.

BAR both  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  do not decrease exponentially with depth (fig. 3b), probably because, unexpectedly, the soil was reworked by man's activity; but the methods used are useful to know the artificial disturbances.

Plots of  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  vs. depth relative to CPR, BAR, CAL and SCA sites are reported in fig. 4. Samples from all these sites cover a depth range much greater than CARP, PENT and LAN sites. In no case do the observed patterns follow a simple exponential decrease with depth, but they are more complex. At CPR and BAR, after 40 cm,  $^{210}\text{Pb}$  is lower than, but well correlated with  $^{226}\text{Ra}$ . At CAL and SCA, the correlation is not so good and, in particular,  $^{210}\text{Pb}$  exceeds  $^{226}\text{Ra}$  in the part of CAL deeper than 2 m and in the 0.9-3.1 m depth range at SCA, indicating that in these layers some Radon emanating from below must have been trapped.

Fractional Radon losses from the soil surfaces at all the sites were calculated using

eq. (4.2) and the plots vs. depth for 5 sites are reported in figs. 5a-e. A general decrease with depth is clear and the patterns are well correlated with the physical characteristics of the soil considering the evidenced layers.

In the SCA site is present an increase in Radon retentivity correlates with a water content increase from 30 to almost 43%, a porosity light increase and a decrease in grain size of almost 10 times (see fig. 5a; the grain size data are not shown). In the deeper layer the Radon is no longer trapped because of the porosity and low water content values and high density and emanation coefficient values.

At LAN the fractional Radon loss is about 0.5 at the surfaces and decreases exponentially with depth down to 90 cm where values sharply drop to less than 0.1 (fig. 5b). This sharp discontinuity occurs at the top of a deeper layer, formed by more compact soil (clay) with smaller grain size, which traps more than 90% of the Radon produced.

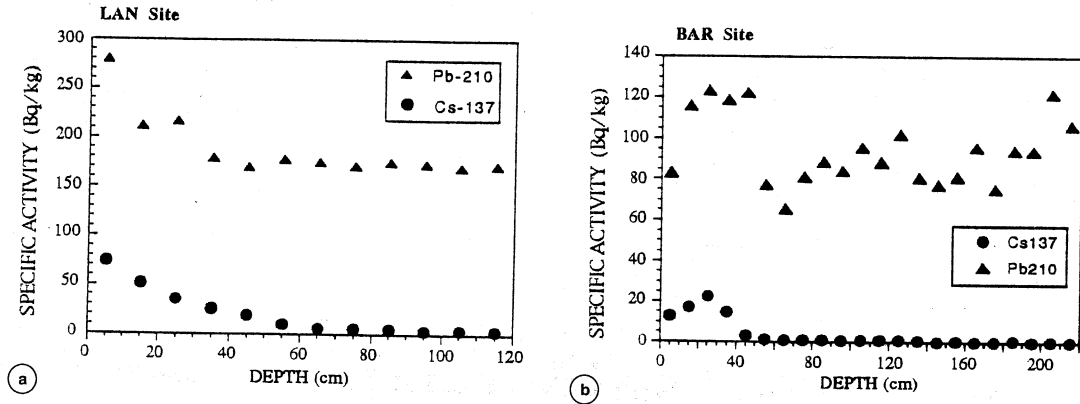


Fig. 3a, b. [ $^{137}\text{Cs}$ ] and [ $^{210}\text{Pb}$ ] vs. depth at LAN and BAR sites. The first site data reflect the natural distribution of two radionuclides, the other data represent a case of artificial disturbance.

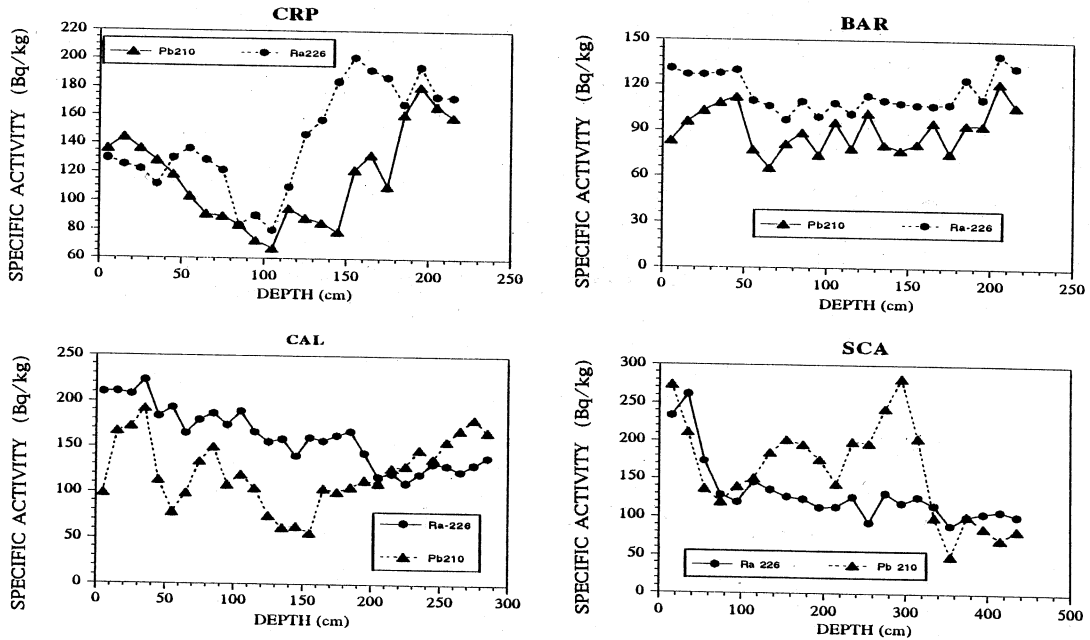
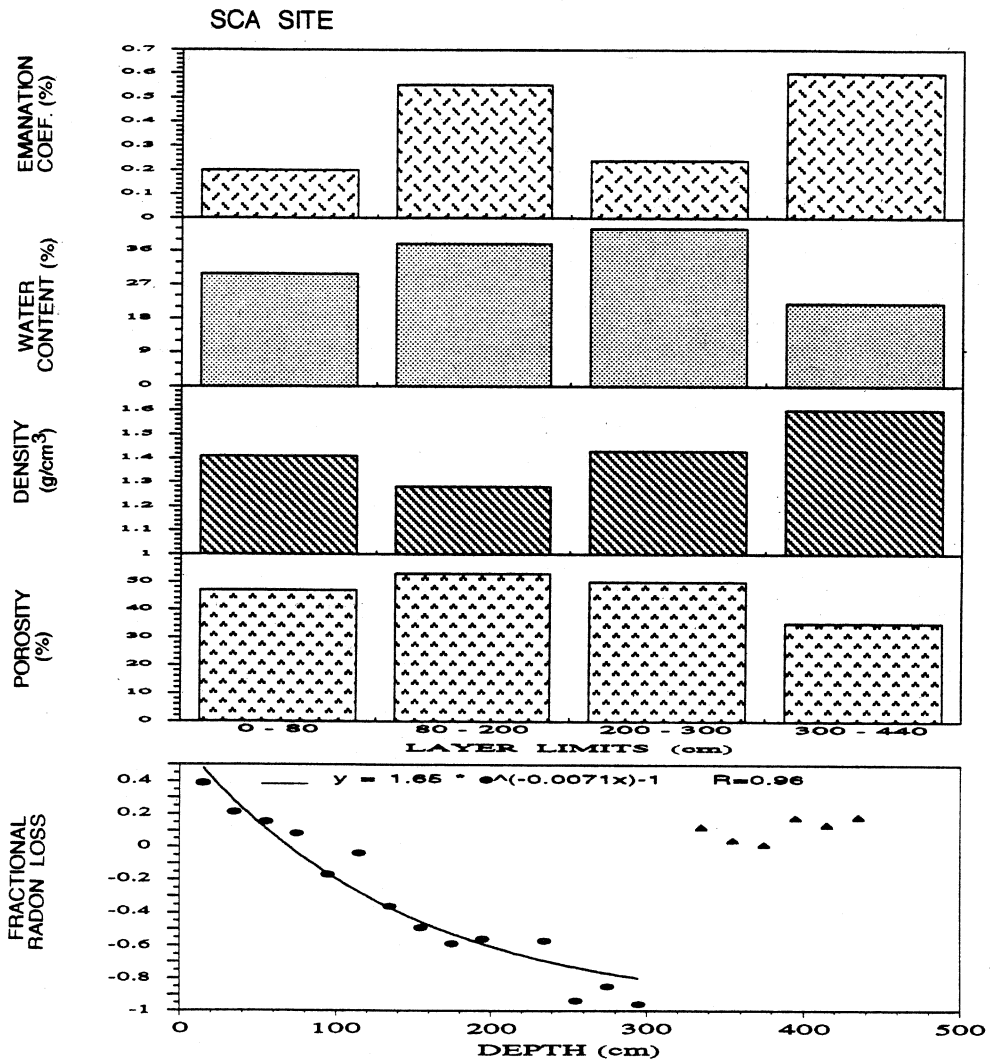


Fig. 4. [ $^{226}\text{Ra}$ ] and [ $^{210}\text{Pb}$ ] vs. depth at CPR, BAR, CAL, SCA sites.

At BAR fractional Radon losses fit a three-layers model, with exponential decrease within each layer and sharp increases at each discontinuity (fig. 5c). The same pattern also appears at CPR and SCA (figs. 5d and 5a). The correlation of fractional Radon loss with soil density

and porosity is contradictory as can easily be observed comparing the patterns at LAN (where the low porosity-high density layer has a higher Radon retention) and at CPR (where Radon retention is lower with decreasing soil porosity and increasing density). The capacity



**Fig. 5a.** Fractional Radon loss against depth at SCA site. Average values of some soil characteristics are also indicated to show the different layers which form each soil. Best-fitting curves of fractional Radon loss in each layer are included.

of a soil to retain Radon appears to be dependent on a combination of different soil parameters, including grain size and water content. This feature is also confirmed at CAL site, which shows a more complicated and less clear pattern. In the deeper layer a diffusional movement is present, extrapolated at the negative val-

ues, which indicate a retention of Radon, probably due to the high emanation coefficient value.

Radon migration parameters have been calculated for the most superficial layer at all the sites, in order to determine the Radon flux at the air-soil interface. The average values obtained at each site are reported in table I. Ema-



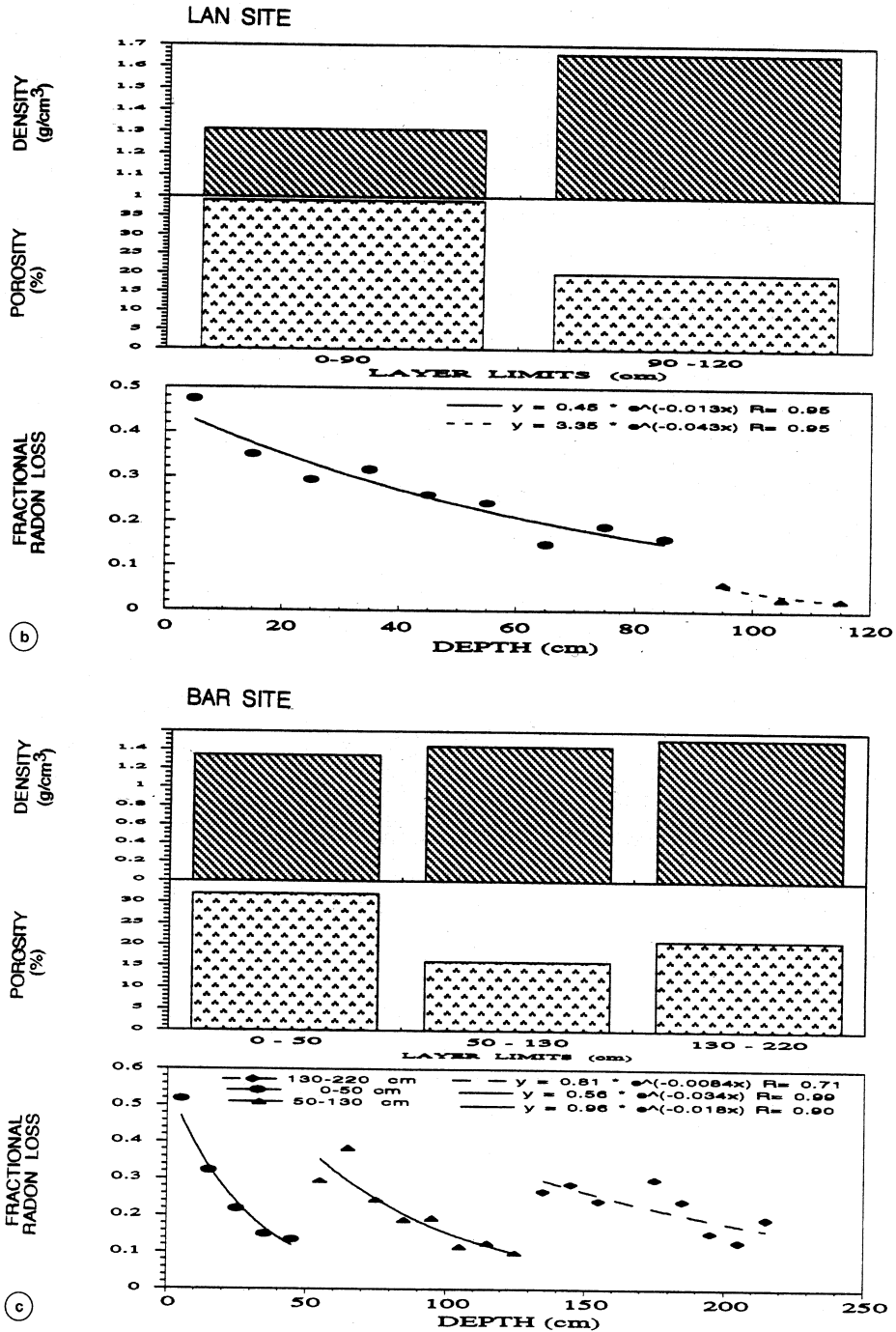


Fig. 5b,c. b) LAN site; c) BAR site. See fig. 5a for explanation.

nation coefficients and average migration lengths are in the range 0.42-0.67 and 23-104 cm respectively. Radon fluxes, calculated by eq. (4.3), range from 0.05 and 0.20 Bq/m<sup>2</sup>s with an average value of 0.11 Bq/m<sup>2</sup>s. Radon flux at the Irno River Valley is higher than fluxes reported by Wilkening *et al.* (1972) and Schery *et al.* (1989) for Australian soils (0.022

Bq/m<sup>2</sup>s) and by Graustein and Turekian (1990) for North America soils (0.040-0.048 Bq/m<sup>2</sup>s). This is due to the occurrence of a significant alkali-potassic magmatic component, produced by the volcanoes of the nearby neapolitan area, whose products are well known to have a very high <sup>226</sup>Ra content (see, for example, Civetta *et al.*, 1970).

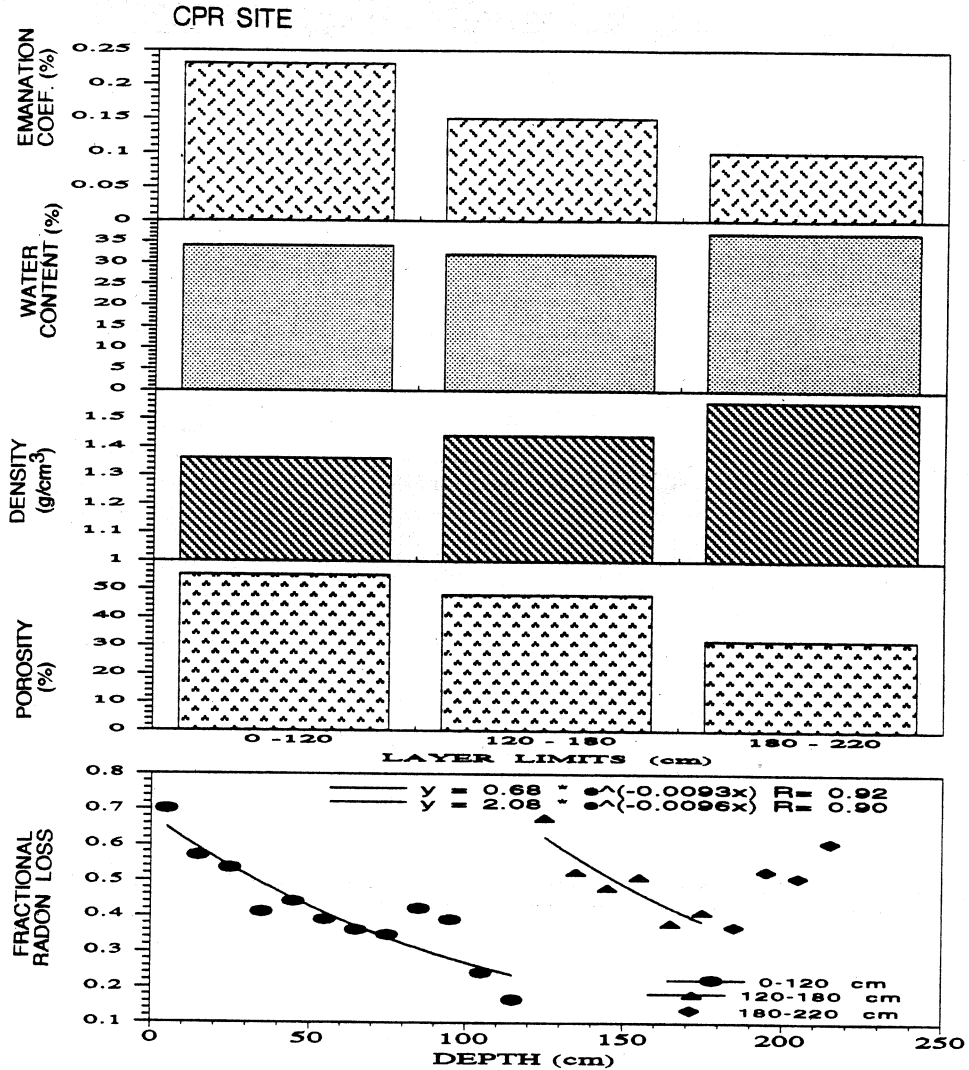


Fig. 5d. CPR site. See fig. 5a for explanation.

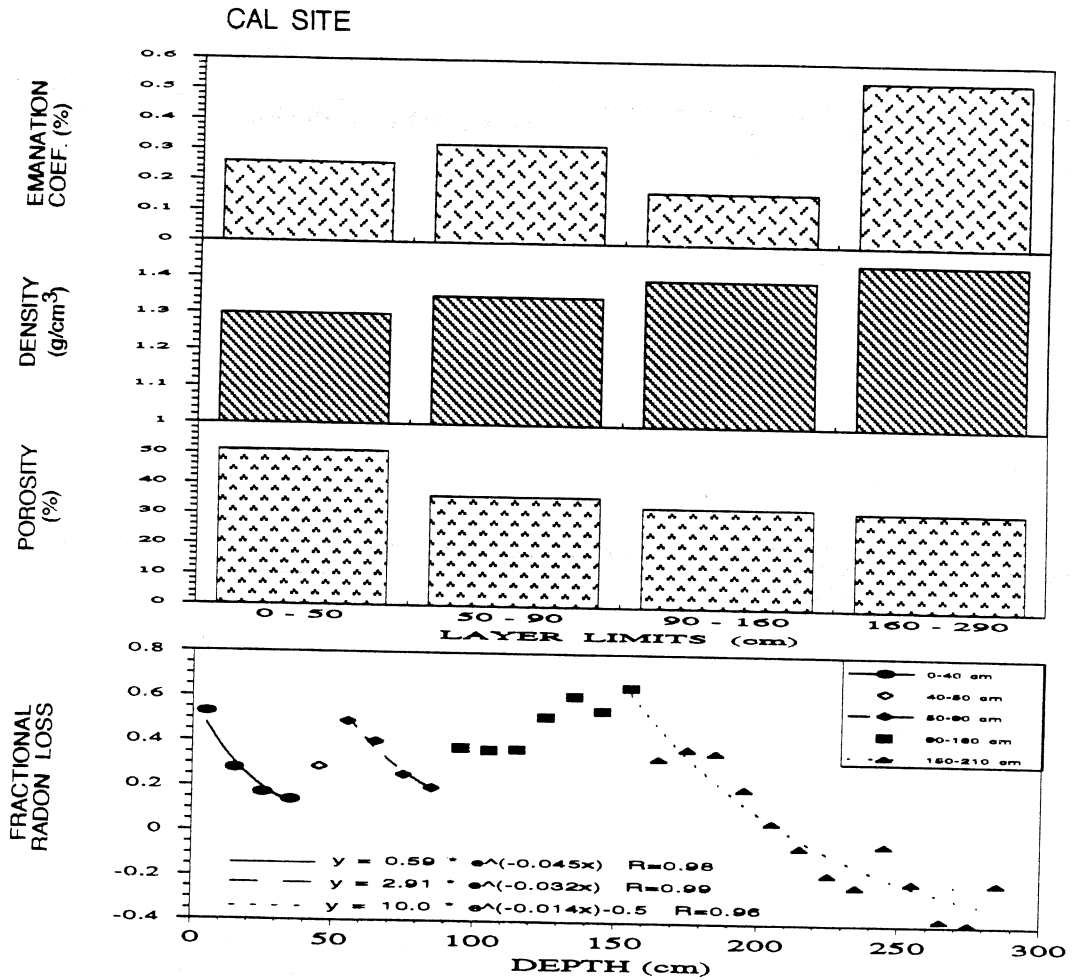


Fig. 5e. CAL site. See fig. 5a for explanation.

## 7. Conclusions

Radon migration in the uppermost layer of the soils covering the Irno River alluvial valley is well described by simple diffusion through a porous medium considering the soil layerings. The exponential decrease in fractional Radon loss with depth is disturbed by changes in the physical parameters of the soil (density, porosity, grain size, water content). The Radon retentivity by the soil does not depend on changes of a single parameter, but rather on

how the changes of various parameters are combined.

When grain size and water content do not change with depth, Radon retentivity increases sharply with decreasing porosity and increasing density, as observed at LAN (fig. 5a). Decrease in grain size and increase in water content are dominant in increasing Radon retentivity, as shown at BAR (fig. 5b). Water rich clay-type layers efficiently trap the Radon emitted from lower layers, as shown at SCA and CAL (figs. 5d and 5e).

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