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## Radon

### 1.1 Physical properties

The gases radon ( $^{222}\text{Rn}$ ) and thoron ( $^{220}\text{Rn}$ ) are formed as progeny of uranium and thorium in rocks and soil. They are emitted from the ground into the atmosphere, where they decay and form daughter products, isotopes of polonium, bismuth and lead, which either remain airborne till they decay, or are deposited in rain and by diffusion to the ground.

Radon and thoron and their decay products are the most important sources of radiation exposure to the general public, contributing on average about half of the total effective dose equivalent received from natural and man-made radioactivity (Clarke & Southwood, 1989).

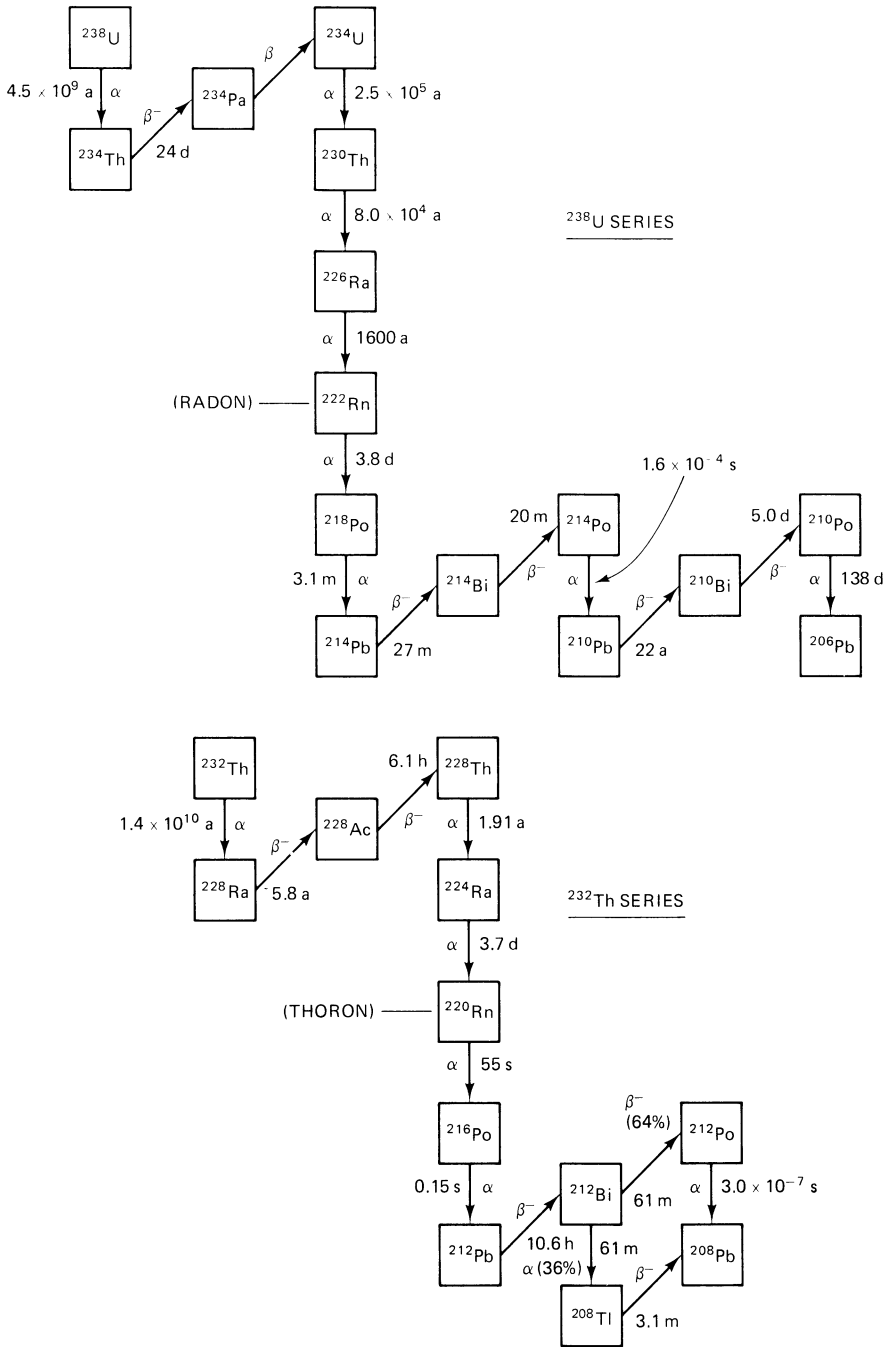
The emanation of a radioactive gas from radium was observed by Madame Curie. In the atmosphere, radon diffuses and mixes with air like any other gas. Rutherford & Brooks (1901) obtained a value  $8 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$  for the diffusivity of radon. Recent determinations are in the range  $1.0$  to  $1.2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$  at N.T.P. (Jost, 1960).

Radon is slightly soluble in water, and obeys Henry's Law. At  $20^\circ\text{C}$  the partition coefficient (amount of radon per litre of water at equilibrium divided by the amount per litre of air) is 0.26. Despite the low solubility, water supplies derived locally from granite and metamorphic rocks can be an important source of airborne radon in dwellings (Nero & Nazaroff, 1984; Hess *et al.*, 1987). Radon is more soluble in fats and organic liquids, and the partition coefficient between air and human fat is about unity at  $37^\circ\text{C}$ .

The radioactive decay schemes of radon and thoron are shown in Fig. 1.1. The old generic nomenclature (RaA, ThB etc.) is now superseded by the isotopic designation ( $^{218}\text{Po}$ ,  $^{212}\text{Pb}$  etc.), but where necessary for clarity the old designation will be added.

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Fig. 1. Radioactive decay series.



## 1.2 Radon in air – methods of measurement

Methods of measuring radon in air, have been reviewed by the US National Council on Radiation Protection & Measurement (1988). The most commonly used methods are as follows.

### (a) *Measurement of activity in ionisation chambers*

The air containing the radon is passed into a chamber. After sufficient time for the decay products down to  $^{214}\text{Po}$  to reach equilibrium with radon, the activity is assessed from the ionisation current. To allow for radioactive contamination in the materials of the chamber, two identical chambers have been used, one filled with the radon-bearing air, the other with aged air, and with the ionisation currents opposed. Using steel chambers of 6.3 l capacity, Hultqvist (1956) measured  $4 \text{ Bq m}^{-3}$  of  $^{222}\text{Rn}$  with 10% accuracy.

An alternative, for low activities of  $^{222}\text{Rn}$ , is to count individual alpha pulses in the ionisation chamber. Kraner *et al.* (1964) used this method to measure exhalation of  $^{222}\text{Rn}$  from the soil. To obtain maximum sensitivity, radon from a large volume of air is adsorbed in activated charcoal, and transferred to an alpha-pulse ionisation chamber in a flow of inert gas.

Ionisation chambers have also been used in a flow-through mode, to give continuous measurement of radon. Israel & Israel (1965) used a very large ionisation chamber, 324 l in volume, for simultaneous measurement of both  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$ . Air was drawn at  $700 \text{ l min}^{-1}$  through a filter into the chamber, and was periodically switched through a 2300-l delay vessel, where the  $^{220}\text{Rn}$  decayed, before entering the chamber. By using a vibrating reed electrometer to measure the ionisation current, sensitivities of about  $0.3 \text{ Bq m}^{-3}$  for both  $^{222}\text{Rn}$  and  $^{224}\text{Rn}$  were obtained. Ionisation chambers are still used for absolute measurements, but for practical purposes they have been superseded by scintillation chambers.

### (b) *Scintillation chambers*

Air with radon is passed into a vessel coated internally with zinc sulphide. Alpha particles from radon in the chamber, and from decay products deposited on the walls, give scintillations which are counted by photomultiplier tubes viewing the chamber through windows (Lucas, 1957). With a chamber of volume 0.1 l, and a counting time of 1 h the detection limit of  $^{222}\text{Rn}$  in air was about  $10 \text{ Bq m}^{-3}$ , but by concentrat-

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ing the radon in charcoal before measurement Lucas (1964) claimed a sensitivity as low as  $4 \text{ mBq m}^{-3}$ .

The scintillation chamber can also be used as a flow-through instrument. Because it takes about 2 h for the alpha decay of  $^{214}\text{Po}$  to come into equilibrium with the  $^{222}\text{Rn}$  in the chamber, calculation is needed to calibrate the instrument. Watnick *et al.* (1986) devised a way round this difficulty by arranging for only the alphas from  $^{218}\text{Po}$  (RaA) to be counted. The decay products produced in the chamber were collected electrostatically on the face of a solid state detector, which had resolution sufficient to separate the alpha particles from  $^{218}\text{Po}$  from those from  $^{214}\text{Po}$ . This enabled a fast response to be obtained, with sensitivity about  $40 \text{ Bq m}^{-3}$  of  $^{222}\text{Rn}$ .

### (c) *Two filter method*

Air is drawn through an efficient filter to remove decay products, and thence through a chamber where fresh decay products are formed. These are collected on an exit filter, and activity is measured either after removal from the chamber, or in situ, usually by alpha scintillation counting. Some decay products are deposited on the walls of the chamber, and their activity can be measured by using a removable lining, or calculated theoretically (Thomas & Le Clare, 1970). If the flow of radon is continued for several hours, the activity of the short-lived products  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  on the exit filter and walls is the same as the activity of  $^{222}\text{Rn}$  in the chamber, and independent of the flow rate through it, so this method can give an absolute calibration.

In applying this method to measurement of  $^{222}\text{Rn}$  in air, Schery *et al.* (1980) used a wide chamber (0.76 m diameter,  $0.6 \text{ m}^{-3}$  volume) to minimise deposition of decay products on the walls. The incoming air was passed through a long pipe in which  $^{220}\text{Rn}$  decayed before entering the chamber. With a flow of 200 to  $300 \text{ l min}^{-1}$ , and counting periods of one or two hours, a sensitivity of  $0.4 \text{ Bq m}^{-3}$  was obtained.

### (d) *Passive radon samplers*

To meet the need to monitor levels of  $^{222}\text{Rn}$  in houses, passive samplers have been developed which measure average concentrations over long periods and do not need power supplies. In the Karlsruhe dosimeter (Urban & Piesch, 1981), a polycarbonate nuclear track detector foil is mounted inside a plastic cup. The mouth of the cup is closed with a filter to allow radon to enter but to exclude decay products. After exposure, the detector foil is etched and the tracks counted optically. This is a

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cheap and reliable method, but the sensitivity is low, and an exposure period of months is needed to measure normal indoor concentrations. However, this has the advantage that seasonal fluctuations are evened out.

In a passive detector developed by the National Radiological Protection Board (Wrixon *et al.*, 1988), the etched pits in the detectors are filled with scintillator fluid. After exposure to radon, the detector is irradiated with an alpha source, and the resulting scintillations counted with a photo-multiplier tube. In this way, track density over  $1 \text{ cm}^2$  of detector can be measured in a few seconds. Passive detectors used in the UK National Survey were sensitive down to  $20 \text{ kBq m}^{-3} \text{ h}$  of accumulated exposure, equivalent to a radon concentration of  $5 \text{ Bq m}^{-3}$  measured over 4000 h exposure.

George (1984) developed a passive sampler in which radon is adsorbed in a canister containing activated charcoal. After exposure, the canister is returned to the laboratory, and the activity of radon, with decay products in equilibrium, measured by gamma spectrometry. The amount of radon adsorbed was found to depend on the relative humidity, and the calibration depended on the amount of water absorbed, as shown by the gain in weight of the canister. Because radon can desorb from charcoal at ambient temperatures, the integrating period is only a few days. The sensitivity is good enough to enable indoor concentrations to be measured with exposure of 72 h.

### 1.3 Emanation of radon and thoron from the soil

Uranium and thorium are widely distributed in the earth's crust. Except in geologically recent sediments, there is equilibrium between parent and daughter nuclides in the decay chains leading from uranium and thorium to the radium isotopes ( $^{224}\text{Ra}$  and  $^{226}\text{Ra}$ ) which are the precursors of  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$ . Table 1.1 shows the mean specific activities of the uranium and thorium chains averaged over a worldwide selection of rock samples (Adams *et al.*, 1959). In 327 samples of

Table 1.1. *Abundance of radioactive elements in rocks*

	Uranium chain		Thorium chain	
	(ppm U)	(Bq kg <sup>-1</sup> )	(ppm Th)	(Bq kg <sup>-1</sup> )
Sedimentary rocks	2.5	33	6.5	26
Igneous rocks	3.5	48	13.5	59

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surface soil from the USA, Myrick *et al.* (1983) found mean activities of 41 and 35 Bq kg<sup>-1</sup> for <sup>226</sup>Ra and <sup>232</sup>Th respectively. There are local variations according to the rock type, and in areas of mineralisation the activities may be up to 1000 times the average.

Typical activities of building materials, such as bricks and concrete, in the UK and USA are in a similar range (20 to 50 Bq kg<sup>-1</sup>) (Nero, 1983), but granite used in older houses in Cornwall has about 100 Bq kg<sup>-1</sup> of radium (O'Riordan *et al.*, 1982). In Sweden, Hultqvist (1956) found about 1000 Bq kg<sup>-1</sup> radium equivalent gamma activity in lightweight concrete made from alum shale.

To measure exhalation of radon a vessel called an accumulator is placed over the soil and sealed to the surface (Wilkening *et al.*, 1972; Keller *et al.*, 1982). To equalise pressures, the vessel is connected to atmosphere or to a bladder by a small vent. The build-up of radon in the accumulator is measured periodically or, in a flow-through arrangement, measurements are made by continuous withdrawal of air to a radon monitor (Schery *et al.*, 1984).

Numerous measurements of <sup>222</sup>Rn exhalation have been reported. Global continental means of 15, 16 and 25 mBq m<sup>-2</sup> s<sup>-1</sup> have been calculated by Israel (1951), Wilkening *et al.* (1972) and Turekian *et al.* (1977), respectively. Higher rates of emission are found in regions of recent tectonic activity, and where the uranium content of the topmost rocks or soil is enhanced.

The concentration of radium per kg in seawater is only about 10<sup>-4</sup> of that in rocks. Exhalation of radon from the ocean has been studied to elucidate gas exchange at the surface (Broecker & Peng, 1974). The emission is typically about 0.04 mBq m<sup>-2</sup> s<sup>-1</sup>, very small compared with land.

The migration of <sup>222</sup>Rn and <sup>220</sup>Rn in the soil, and the factors affecting release to atmosphere have been reviewed by Tanner (1964). The energy imparted by the alpha decay of their precursors causes radon atoms to recoil along a track of about 0.03 μm in minerals and about 80 μm in air. Despite the very short range, it seems that release depends more on recoil than on diffusion within the mineral crystal. Emanation to the interstitial air may be primarily from <sup>226</sup>Ra or <sup>224</sup>Ra on the surface of the crystals.

The emanating power or coefficient of rocks or soil is defined as the proportion of the radon activity per unit bulk volume which is in the interstitial gas. The emanating coefficients of rocks and soils vary greatly. Barretto *et al.* (1972) found values ranging from 0.01 to 0.26 for

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rocks and from 0.1 to 0.55 for soils. Up to a certain limit, moisture in soil may increase the emanating power. Recoiling radon atoms collide with water molecules in the pore space between grains (Standen *et al.*, 1984), but remain in the interstitial gas as the solubility is low.

Radon moves upwards in soil partly by molecular diffusion in soil gas and partly by bulk flow caused by changes in air pressure at the surface. The diffusion flux can be calculated, if it is assumed that the radioactivity, porosity and density of the soil are independent of depth and that lateral movement of radon can be neglected.

If  $\rho_s$  ( $\text{kg m}^{-3}$ ) is the bulk soil density,  $A_R$  ( $\text{Bq kg}^{-1}$ ) the specific activity of radium in the soil,  $\alpha$  the emanating coefficient, and  $\lambda$  ( $\text{s}^{-1}$ ) the decay constant of radon, then  $\rho_s A_R \alpha$  atoms, or  $\rho_s A_R \alpha \lambda$  Bq of radon, enter the interstitial air per  $\text{m}^3$  of soil volume per second. At depth in the soil, the rates of entry and radioactive decay of radon are equal, so its activity in interstitial air is

$$\chi_\infty = \rho_s A_R \alpha \text{ Bq m}^{-3} \text{ of soil} \quad (1.1)$$

Near the surface, radon diffuses upwards. If  $\chi(z)$  is the concentration at depth  $z$  and  $D_e$  the effective diffusion coefficient of radon in soil:

$$\rho_s A_R \alpha \lambda - \lambda \chi(z) + D_e \frac{d^2 \chi(z)}{dz^2} = 0 \quad (1.2)$$

Note that  $\chi$  is the concentration of radon in the pore spaces per  $\text{m}^3$  of soil. The concentration per  $\text{m}^3$  of interstitial air in the soil is  $\chi/\varepsilon$ , where  $\varepsilon$  is the porosity of the soil. Equation (1.2) gives

$$\chi(z) = \chi_\infty [1 - \exp(-z/L)] \quad (1.3)$$

where

$$L = (D_e/\lambda)^{\frac{1}{2}} \quad (1.4)$$

$L$  is termed the diffusion length, and is a measure of the depth of soil from which diffusion of radon is effective. The flux of radon at the surface is

$$Q = D_e \left( \frac{d\chi(z)}{dz} \right)_{z=0} \quad (1.5)$$

$$= \chi_\infty D_e L^{-1} \quad (1.6)$$

$$= \chi_\infty \lambda L \quad (1.7)$$

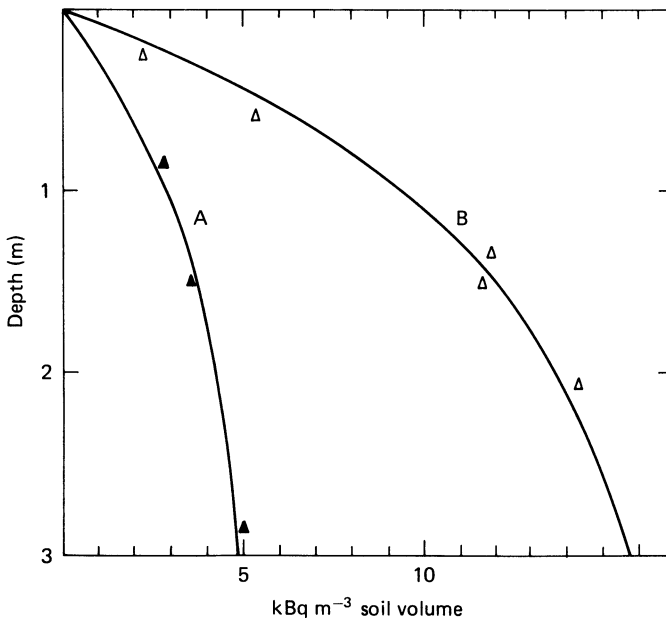
by substituting  $D_e$  from (1.4).

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Kraner *et al.* (1964) at Yucca Flat, Nevada, and Schery *et al.* (1984) at Socorro, New Mexico, drew interstitial air from soil at various depths through sampling tubes and measured  $^{222}\text{Rn}$  in it. Figure 1.2 shows their results converted to  $\chi(z)$  by multiplying by the appropriate values of  $\varepsilon$  (0.25 at Yucca Flat and 0.35 at Socorro). The curves A and B in Fig. 1.2 are fitted using (1.3) with parameters given in Table 1.2. As it happens, the same value  $L = 1.3$  m gives a good fit to both sets of data. Since  $\lambda$  for  $^{222}\text{Rn}$  is  $2.1 \times 10^{-6} \text{ s}^{-1}$ , the corresponding value of  $D_e$  from (1.4) is  $3.5 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ . The exhalation fluxes, deduced from the curves in Fig. 1.2 using (1.7) are 15 and 48  $\text{mBq m}^{-2} \text{ s}^{-1}$  at Yucca Flat and Socorro respectively.

Kraner *et al.* and Schery *et al.* also measured  $Q$  directly, by placing accumulator chambers over the ground. Changes in barometric pressure were found to affect  $Q$  and also  $\chi$  at shallow depths. On days of light winds, Kraner *et al.* found that the flux averaged  $19 \text{ mBq m}^{-2} \text{ s}^{-1}$ , in fair agreement with the value deduced from the gradient in soil, but on windy days the flux averaged  $26 \text{ mBq m}^{-2} \text{ s}^{-1}$ . Over periods of stable barometric pressure, Schery *et al.* at Socorro measured an average  $Q$  of

Fig. 1.2. Variation with depth in soil of interstitial radon.  $\blacktriangle$ , Kraner *et al.*, 1964;  $\triangle$ , Schery *et al.*, 1984. Curves A, B are equation (1.3) with  $L = 1.3$  m.





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Table 1.2. *Exhalation of radon from soil*

Location	Yucca Flat	Socorro
Soil type	Weathered tuff	Gravelly sandy loam
Porosity	0.25	0.35
$\chi_\infty$ , kBq per m <sup>3</sup> soil	5.4	17.5
$L$ , m	1.3	1.3
$Q$ , mBq m <sup>-2</sup> s <sup>-1</sup>		
calculated	15	48
measured	19	34

34 mBq m<sup>-2</sup> s<sup>-1</sup>. In the short term,  $Q$  varied by a factor 2 due to pressure effects, but Schery *et al.* found that the time averaged exhalation was broadly consistent with the diffusion model.

Both Yucca Flat and Socorro are in arid districts and have porous soils. Lower values of  $D_e$  are found in clayey soils (Tanner, 1964, 1980).

As an order of magnitude calculation for average conditions,  $A_R$  can be taken as 30 Bq kg<sup>-1</sup> (Table 1.1). With  $\rho_s = 1.5 \times 10^3$  kg m<sup>-3</sup> and  $\alpha = 0.25$ , equation (1.1) gives  $\chi_\infty = 11$  kBq m<sup>-3</sup>, intermediate between the values for Yucca Flat and Socorro in Table 1.2. If  $L$  is taken as 1 m, equation (1.7) gives  $Q = 23$  mBq m<sup>-2</sup> s<sup>-1</sup>, which is near the reported global average.

Radon entering buildings comes partly from the underlying soil and partly from building materials. The emanation coefficients and diffusion length in brick and concrete are generally somewhat less than in soil (Ingersoll, 1983), and it is thought that radon in most domestic buildings comes from the soil except where building materials have exceptionally high radium content. Pressure effects are more important in buildings than in the open air because there is usually a small persistent negative pressure in buildings relative to the open air (Nero & Nazaroff, 1984). Pressure variations are a dominant factor in the entry of <sup>222</sup>Rn into mine workings, and reductions in concentration have been achieved by maintaining positive air pressure during working, and negative pressure during non-working hours.

The decay constant of <sup>220</sup>Rn is 6000 times greater than that of <sup>222</sup>Rn, so from (1.4)  $L$  is about 80 times less, that is only about 1 cm. The fraction of <sup>220</sup>Rn atoms which escape from the rock crystals to the interstitial air is apparently about the same as for <sup>222</sup>Rn, and since the specific activities of the thorium and radium chains are similar, equation (1.6) implies that the emanation of <sup>222</sup>Rn should be 80 times greater

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measured in activity, but 80 times less measured in atoms  $\text{m}^{-2} \text{s}^{-1}$ , than the emanation of  $^{220}\text{Rn}$ . Zarcone *et al.* (1986), in a mineralised area of New Mexico, measured exhalation rates of 3.6 and  $0.067 \text{ Bq m}^{-2} \text{ s}^{-1}$  for  $^{220}\text{Rn}$  and  $^{222}\text{Rn}$ , a ratio of 54 : 1.

### 1.4 Radon in air – variations in space and time

Very many measurements of radon in air have been made. Table 1.3 compares results of old measurements (Satterly, 1908, radon absorbed in charcoal and transferred to ionisation chamber) and recent measurement (Keller & Folkerts, 1984,  $^{218}\text{Po}$  collected from a chamber followed by alpha spectroscopy). At 15 locations in the UK, Wrixon *et al.* (1988), using passive dosimeters out of doors, found a mean of  $3.4 \text{ Bq m}^{-3}$ , almost the same as Satterly's result obtained at Cambridge 80 years previously. Year round measurements from four locations in the USA gave average  $^{222}\text{Rn}$  in the range 8–12  $\text{Bq m}^{-3}$  (Gesell, 1983). Somewhat lower mean levels would be expected in the UK than in USA or Germany, owing to the influence of oceanic air. Most stations show higher mean levels in winter than summer, because vertical dispersion is better in summer. For the same reason, levels are generally higher at night. Over the oceans, and over snow in the Arctic and Antarctic, the radon concentration is about two orders of magnitude lower than over land (Israel, 1951; Lockhart, 1960).

The concentration of radon decreases with height, the gradient being determined by the vertical diffusivity of the atmosphere. Jacobi & André (1963) calculated the gradient by solving numerically the equation

$$\frac{d}{dz} \left( K(z) \frac{d\chi}{dz} \right) - \lambda\chi = 0 \quad (1.8)$$

where  $K(z)$  is the eddy diffusivity at height  $z$ . They assumed five characteristic profiles of  $K(z)$ , corresponding to five categories of

Table 1.3. *Radon in air near ground level*

Location	No. of measurements	$^{222}\text{Rn}$ ( $\text{Bq m}^{-3}$ )			Reference
		Mean	Highest	Lowest	
Cambridge, UK	58	3.9	13	1.3	Satterly (1908)
Saarland, FRG	101	6.7	17	1.1	Keller & Folkerts (1984)