${f 2.}~~^{222}{f Rn}$ and $^{220}{f Rn}$ fundamentals

Being an inert gas, the main characteristic of ²²²Rn and ²²⁰Rn among other natural radioelements is the fact that their behaviour is not affected by chemical processes. In addition, their concentration levels depend strongly on geological and geophysical conditions, as well as on atmospheric influences such as barometric pressure and rainfall. Formed as a result of the natural radioactive series in the earth's crust they are free to move through soil pores and rock fractures; then to escape into the atmosphere. ²²²Rn and/or ²²⁰Rn exhaled from the earth's surface into the free atmosphere is rapidly dispersed and diluted by natural convection and turbulence. When a dwelling is present, they may migrate into this structure and accumulate indoors in sufficient quantities to pose a health hazard.

2.1. Physical and chemical properties

A characteristic common to all of the natural radioactive series, unlike the artificially produced neptunium series, is the existence of radon isotopes. This element, with an atomic number of 86, is a colorless, odorless, tasteless and radioactive noble gas that generally lacks activity toward other chemical agents. It is the heaviest member of the rare gas group (\sim 100 times heavier than hydrogen and \sim 7.5 times heavier than air). When cooled below its freezing point, radon exhibits a brilliant phosphorescence that becomes yellow at lower temperatures and orange-red at the temperature of the liquid air (CRC, 2001). The electronic structure of its atom suggests very limited chemical reactivity. However, according to its relatively low first-ionisation potential of 10.7 eV some interactions might be possible. Radon can react with fluorine to produce radon fluoride (RnF₄) and when its atoms are found within crystal lattices, of certain hydrogenate compounds, clathrates are formed. Radon is readily absorbed on charcoal, silica gel and similar substances. This property serves to separate it from other gases by collecting it on activated charcoal cooled

¹Clathrates — from a Latin word meaning "enclosed by a lattice" — are chemical substances in which the molecules of one compound, called the host, form a cagelike crystalline lattice within which there are open spaces that may be occupied by molecules of a second compound or element, called the guest, although no traditional valence bonds appear in the formation of such substances.

to the temperature of solid CO₂ (-78.2 °C). Radon is released from charcoal by heating to 350 °C. Another property of radon is its solubility in various liquids such as water or natural gas which may transport it over large distances through the soil, so that in special cases, when they are radon-rich elements, their contribution to the total indoor air concentration might be important. A detailed list of the physical properties of radon is given in Table 2.1.

Table 2.1. Physical properties of radon.

Density at 1 atm pressure and 0 °C	$9.73 \mathrm{~g~L^{-1}}$
Boiling point at 1 atm pressure	-62 °C
Density of liquid at normal boiling point	4.4 g cm^{-3}
Diffusion coefficient in air	$0.1 \text{ cm}^2 \text{ s}^{-1}$
Diffusion coefficient in water	$10^{-5} \text{ cm}^2 \text{ s}^{-1}$
Viscosity at 1 atm pressure and 20 $^{o}\mathrm{C}$	0.229 poise
Solubility in water at 1 atm pressure and 20 $^{o}\mathrm{C}$	$230 \text{ cm}^3 \text{ kg}^{-1}$
Solubility in various liquids at 1 atm pressure and 18 $^{o}\mathrm{C}$	
glycerine	$0.2~{\rm cm^3~kg^{-1}}$
ethyl alcohol	$7.4 \text{ cm}^3 \text{ kg}^{-1}$
petroleum (liquid paraffin)	$9.2 \text{ cm}^3 \text{ kg}^{-1}$
toluene	$13.2~{\rm cm^3~kg^{-1}}$
carbon disulfide	$23.1 \text{ cm}^3 \text{ kg}^{-1}$
olive oil	$29.0 \text{ cm}^3 \text{ kg}^{-1}$

2.2. Radiometric properties

The radiometric properties of radon isotopes and their daughters are used for detection; these properties also confer the potential for induction of biological effects. Since radon is produced continuously from the α-decay of radium isotopes in rocks and minerals, there are three natural isotopes of this radioelement in the environment — ²²²Rn, ²²⁰Rn, and ²¹⁹Rn (actinon) originated from the so-called decay series of ²³⁸U (uranium), ²³²Th (thorium), and ²³⁵U (actinium), respectively. Detailed listing of these natural radioactive decay series are shown in Figures 2.1, 2.2, and 2.3². Radon's importance as an environment source of radiation depends principally on the local concentrations of the parent — radium — isotopes, on the physical characteristics of its source medium, and on its mean life.

Actinon, because of its half-life of only 3.96 s, has limited capacity to migrate into air environment. Likewise, due to the lower relative abundance of 235 U by weight in

²The energies of α -emissions and the branching ratios were obtained using the JEF-PC 2.0 computer package for viewing evaluated and experimental nuclear data developed jointly by the Nuclear Energy Agency (NEA) and the University of Birmingham, England.

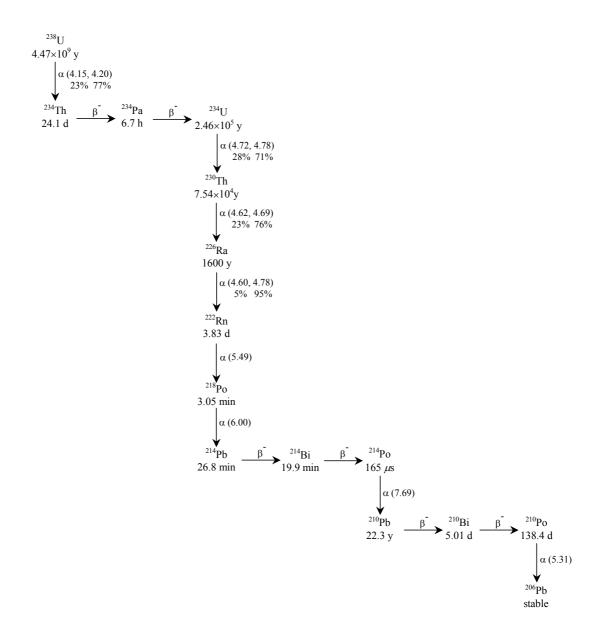


Figure 2.1. Decay diagram of 238 U series with the half-life of each radionuclide and the energies of α -emissions expressed in MeV.

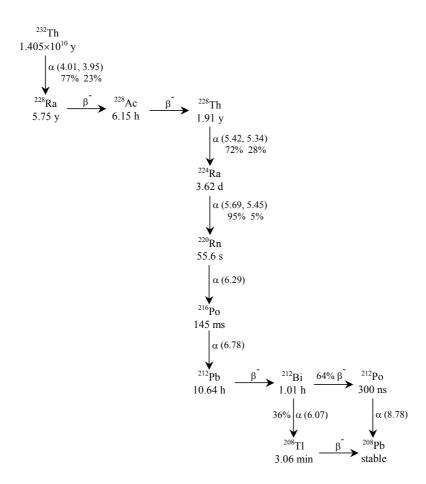


Figure 2.2. Decay diagram of 232 Th series with the half-life of each radionuclide and the energies of α -emissions expressed in MeV.

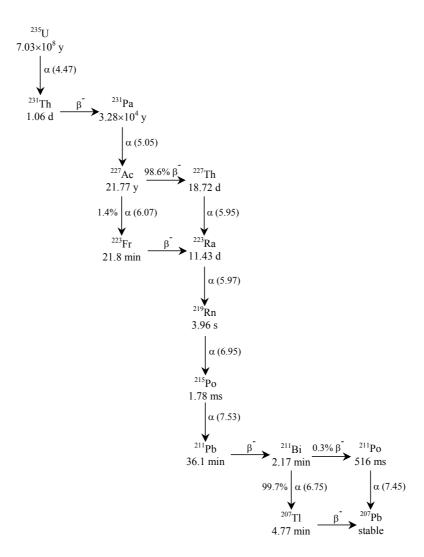


Figure 2.3. Decay diagram of 235 U series with the half-life of each radionuclide and the energies of α -emissions expressed in MeV.

natural uranium — $\sim 0.711\%$ (CRC, 2001) — this radioelement is extremely rare in the atmosphere and can generally be neglected in radon dose considerations. Even though thorium is generally more abundant than uranium in the earth's crust, its probability for decay is smaller; hence, the production rates of 222 Rn and 220 Rn in the soil are roughly the same. As the half-life of 220 Rn is no greater than one minute, much of its atoms decay before reaching the earth's surface and its mean diffusion length, which is proportional to the square root of the mean life τ , is expected to be a factor of ~ 77 lower to that of 222 Rn. Afterwards, only a small thickness of soil or building material serves as an effective source for 220 Rn entering the atmospheric environment, indoors or outdoors. The 222 Rn is by far the dominant radionuclide in indoor air and constitutes the major concern as a health hazard in the environment. Given an uniform entry rate from soil and building materials, the only removal process decreasing the indoor concentration of 220 Rn is by decaying, while 222 Rn may also be reduced by ventilation (see Section 2.5.4). As a result of that, the possible health significance of 220 Rn is not to be ignored, and, indeed, its concentration must be taken into account.

Once in the air atmosphere, the 222 Rn and 220 Rn atoms decay producing isotopes of polonium, lead, and bismuth — also thallium for 220 Rn. These elements are heavy metals chemically very active, which may exist briefly as ions and/or free atoms before forming molecules in condensed phase or attached to airborne dust particles, typically to those with a sub-micron range of sizes, forming radioactive aerosols. A variable proportion of airborne 222 Rn and 220 Rn daughters remains unattached and is referred as the airborne-unattached fraction. This fraction may be inhaled and deposited in the respiratory tract, in which they release all their α -emissions. The particle size distribution of the aerosol-attached fraction in the inhaled air also influences the dose to the airways, because particles of different sizes deposit preferentially in different areas of the respiratory tract.

²²²Rn decay products are divided into two groups, namely, the short-lived (²¹⁸Po, ²¹⁴Pb, ²¹⁴Bi, and ²¹⁴Po) and the long-lived (²¹⁰Po, ²¹⁰Pb, and ²¹⁰Bi) daughters. Since the longest-lived element of the first group has a half-life of less than 27 min, the whole sequence of decays can be completed before the human clearance processes can sweep them away. The long-lived ²²²Rn progeny contributes relatively little to lung exposure because the first nuclide, ²¹⁰Pb, of this group has a large half-life (22.3 y) so that is utterly removed from the body before decaying. The case of ²²⁰Rn is somewhat different, firstly there is no long-lived group of its daughters and secondly the most important radionuclide in its chain is the lead isotope ²¹²Pb, which has a relatively long half-life of 10.64 h. In this way, a considerable fraction of this radionuclide deposited in the bronchial epithelium can be absorbed into blood; so that it may be carried to other organs and may produce a large biological impact.

2.3. Radioactive quantities and units of measurement

In order to estimate the dose of α -energy from inhaled short-lived $^{222}\mathrm{Rn}$ and $^{220}\mathrm{Rn}$ progeny in private homes and in workplaces, it is necessary to know how these radionuclides move in the atmosphere. The special quantities and units used in this study are taken from the definitions given in the International Commission on Radiological Protection (ICRP) publications (ICRP, 1987; ICRP, 1994).

Amounts of radioactive material can be described in units of decays per unit time or activity rather than as the mass of contaminant. The activity, A, is the number of decay events per unit time and is calculated as

$$A = \lambda N \tag{2.1}$$

where λ (s⁻¹) is the probability of decay per unit time of the nucleus of a particular atom $-\lambda = \frac{\ln(2)}{T}$, T being the half-life of the isotope — and N is the total number of the atoms present in the sample. The International System (SI) unit of activity is the Becquerel (Bq), which corresponds to one decay per second. For airborne concentrations of 222 Rn, 220 Rn and their progeny, the amount of activity per unit volume — $C_i = \frac{A_i}{V}$, with a proper unit of Bq m⁻³ — is ordinarily used. The potential α -energy per atom, E_{pi} , of the short-lived daughter i of 222 Rn or 220 Rn is the sum of all α -energies emitted during its decay (see Figures 2.1 and 2.2) to 210 Pb or 208 Pb, respectively. When there are N_i atoms of this daughter in the airborne, its total potential α -energy becomes

$$N_i E_{\mathrm{p}i} = \frac{A_i E_{\mathrm{p}i}}{\lambda_i} \tag{2.2}$$

where λ_i and A_i are, respectively, the decay constant and the activity of the daughter i of 222 Rn or 220 Rn.

Table 2.2 lists the potential α -energy per atom and per unit of activity for each of the short-lived 222 Rn and 220 Rn progeny (ICRP, 1987). The potential α -energy concentration of any mixture, $C_{\rm p}$, of short-lived 222 Rn and/or 220 Rn daughters is the sum of the potential α -energy of all daughter atoms present per unit volume of air and is given by

$$C_{\rm p} = \sum_{i=1}^{4} C_{\rm p}i = \sum_{i=1}^{4} \frac{C_i E_{\rm p}i}{\lambda_i}$$
 (2.3)

This quantity is expressed in SI-units as J m⁻³ (1 J = 6.24×10^{12} MeV). Traditionally, the special unit Working Level (WL) was often used for C_p . The WL corresponds to the potential α -energy concentration of the short-lived ²²²Rn decay products, which are in secular equilibrium with their noble gas parent at a concentration of 3700 Bq m⁻³. For

 220 Rn daughters in radioactive equilibrium with their parent nuclide, 1 WL corresponds to a 220 Rn concentration of 275 Bq m $^{-3}$. Then, we have

$$C_{\rm p}^{\rm Rn} ({\rm WL}) = \frac{C_{\rm eq}^{\rm Rn} ({\rm Bq~m}^{-3})}{3700} \text{ and } C_{\rm p}^{\rm Tn} ({\rm WL}) = \frac{C_{\rm eq}^{\rm Tn} ({\rm Bq~m}^{-3})}{275}$$
 (2.4)

where the superscripts Rn and Tn stand for 222 Rn and 220 Rn case, respectively.

Table 2.2. Decay constants, λ_i , and potential α -energy per atom, $E_{p,i}$, and per Bq of activity, $\frac{E_{p,i}}{\lambda_i}$, of the short-lived 222 Rn and 220 Rn daughters.

		λ_i Pote			l α -energy	
Radionuclide	i	h^{-1}	s^{-1}	$E_{pi} \text{ (MeV)}$	$\frac{E_{\mathrm{p}i}}{\lambda_i}$ (MeV s)	$k_{\mathrm pi}^*$
-222Rn	0	7.54×10^{-3}	2.10×10^{-6}	19.18	9.13×10^{6}	_
218 Po	1	13.64	3.78×10^{-3}	13.69	36.22×10^2	0.105
$^{214}\mathrm{Pb}$	2	1.55	4.31×10^{-4}	7.69	17.84×10^3	0.515
$^{214}\mathrm{Bi}$	3	2.10	5.83×10^{-4}	7.69	13.19×10^3	0.380
214 Po	4	15.12×10^6	42.00×10^2	7.69	$1.83{ imes}10^{-3}$	$5.28{ imes}10^{-8}$
$^{220}\mathrm{Rn}$	0	44.88	1.25×10^{-2}	20.87	16.70×10^2	_
216 Po	1	17.21×10^3	4.78	15.58	3.26	6.91×10^{-6}
$^{212}\mathrm{Pb}$	2	6.51×10^{-2}	1.81×10^{-5}	7.80	43.09×10^4	0.914
$^{212}\mathrm{Bi}$	3	0.69	1.92×10^{-4}	7.80	40.62×10^3	0.086
²¹² Po	4	83.17×10^8	23.10×10^{5}	8.78	3.80×10^{-6}	8.06×10^{-12}

 $[*]k_{pi} = \frac{\frac{E_{pi}}{\lambda_i}}{\sum_{i=1}^{4} \frac{E_{pi}}{\lambda_i}} \text{ where } \sum_{i=1}^{4} k_{pi} = 1$

The indoor 222 Rn and 220 Rn decay products are not in radioactive equilibrium with their parent nuclides, even under steady-state conditions³. The reason for this is the removal process from the indoor air by ventilation and/or by deposition on any available surface (walls, floor, ceilings, and furniture). Therefore, the potential α -energy concentration in air is expressed in terms of the so-called equilibrium-equivalent concentration, $C_{\rm eq}$, of their parent nuclide (222 Rn or 220 Rn). The equilibrium-equivalent concentration, $C_{\rm eq}$, is a concentration of 222 Rn (220 Rn) assumed to be in radioactive equilibrium with its short-lived daughters present in the considered air volume that would give the same potential α -energy concentration, $C_{\rm p}$, as the actual non-equilibrium mixture; so that, we can write

³The steady-state approach is usefull and not unreasonable as in most cases of human exposure we are concerned with effects of long-term average concentrations rather than those arising from short-term variations.

$$C_{\rm p} = C_{\rm eq} \sum_{i=1}^{4} \frac{E_{\rm p}i}{\lambda_i} \tag{2.5}$$

where the conversion factor $(\sum_{i=1}^{4} \frac{E_{pi}}{\lambda_i})$ corresponds to 5.6×10^{-9} J Bq⁻¹ in the case of ²²²Rn progeny and 7.6×10^{-9} J Bq⁻¹ for ²²⁰Rn progeny (see Table 2.2). This definition, taking into account the Equation (2.3), leads to the following relationship for ²²²Rn decay products

$$C_{\text{eq}}^{\text{Rn}} = \frac{\sum_{i=1}^{4} \frac{C_i E_{\text{p}i}}{\lambda_i}}{\sum_{i=1}^{4} \frac{E_{\text{p}i}}{\lambda_i}} = \sum_{i=1}^{4} k_{\text{p}i} C_i = 0.105 C_1 + 0.515 C_2 + 0.380 C_3$$
 (2.6)

The contribution of the 214 Po concentration to this expression is in any case negligible ($k_{\rm p4} \simeq 0$) due to its short half-life (165 μ s), causing very few atoms of this radionuclide to be present in air as compared with other short-lived decay products. The analogous expression of $C_{\rm eq}$ for the 220 Rn series is

$$C_{\text{eq}}^{\text{Tn}} = 0.914 \, C_2 + 0.086 \, C_3 \tag{2.7}$$

The short-lived ²²⁰Rn daughters, ²¹⁶Po and ²¹²Po, do not contribute directly to the expression (2.7), also because of their short half-lives (see Figure 2.2).

In order to characterise the actual non-equilibrium between the airborne daughter mixture and their parent nuclide in terms of potential α -energy, the equilibrium factor F is defined as the ratio of the $C_{\rm eq}^{\rm Rn}$ ($C_{\rm eq}^{\rm Tn}$) to the $^{222}{\rm Rn}$ ($^{220}{\rm Rn}$) concentration in air. F=1 for complete radioactive equilibrium between $^{222}{\rm Rn}$ ($^{220}{\rm Rn}$) and its progeny, and F=0 for pure gas with no daughters.

Another important quantity in the radon field is the disequilibrium degree between the airborne concentration of each daughter i to that of its parent (222 Rn or 220 Rn), which is characterised by the concentration ratio

$$f_i = \frac{C_i}{C_0} = \frac{C_i^{\mathrm{u}}}{C_0} + \frac{C_i^{\mathrm{a}}}{C_0} = f_i^{\mathrm{u}} + f_i^{\mathrm{a}}$$
(2.8)

where C_i^{u} and C_i^{a} are, respectively, the portions of its atoms that are unattached and attached to the airborne aerosol particles.

To estimate the radiation effective dose, the potential α -energy intake of an individual by inhalation of ^{222}Rn and/or ^{220}Rn daughter mixture during a given period of time must be defined. If v (m³ h⁻¹) is the mean breathing rate during this period, the potential α -energy intake, I_p , is related to the potential α -energy concentration, C_p , by the equation

$$I_{\rm p} = vC_{\rm p}t_{\rm e} = v\chi_{\rm p} \tag{2.9}$$

where $t_{\rm e}$ (h) is the exposure time and $\chi_{\rm p}$ (J m⁻³ h) is the potential α -energy exposure which is defined as the time-integral over the potential α -energy concentration of any ²²²Rn and/or ²²⁰Rn daughter mixture. For this purpose, it is adequate to use an occupancy time of ~2000 hours per year (170 h per month) at work and of ~7000 hours (580 h per month) for the members of the public in private homes (ICRP, 1994).

A former unit of $\chi_{\rm p}$ is the so-called Working Level Month (WLM), with 1 WLM corresponding to a cumulative exposure of 1 WL during the reference working period of 1 month. As 1 WL corresponds to an equivalent-equilibrium concentration of 3700 Bq m⁻³ for ²²²Rn (275 Bq m⁻³ for ²²⁰Rn), a potential α -energy exposure of 1 WLM is associated with an equilibrium-equivalent activity exposure of 6.3×10^5 Bq m⁻³ h for ²²²Rn progeny at work and 2.15×10^6 Bq m⁻³ h in private homes (respectively, 4.68×10^4 Bq m⁻³ h and 1.6×10^5 Bq m⁻³ h for ²²⁰Rn progeny).

Since the present work utilises the SI accepted units and in some references traditional units could be found, Table 2.3 gives the conversion factors between these units.

	-	-
Parameter	Traditional unit	Conversion for SI-unit
Activity	Ci	$3.7 \times 10^{10} \text{ Bq}$
Concentration	$ m pCi~L^{-1}$	$37~\mathrm{Bq~m^{-3}}$
$C_{ m p}$	WL	$2.08 \times 10^{-5} \text{ J m}^{-3}$
Exposure	$pCi L^{-1} d$	$0.89 \text{ kBq m}^{-3} \text{ h}$

Table 2.3. Traditional units and their equivalents in the SI accepted units.

Relationships between exposure and dose

The radiation effective dose from indoor exposure to 222 Rn, 220 Rn and their progeny can be derived from a measurement of the potential α -energy concentration combined with Dose Conversion Factor (DCF). The dose determination over all epithetical cells in the tracheo-bronchial regions is extremely complex and is dependent on both biological and non-biological factors, including the physical characteristics and the amount of the inhaled air, breathing patterns, mucociliary clearance, particle deposition, airway geometry and location of the target cells. As the effective dose of α -energy delivered to target cells in the lungs cannot be measured directly, modelling approaches are used. Many dosimetric models of respiratory tract have been developed recently showing a wide range of calculated dose conversion factors (NRC, 1991; ICRP, 1994; NRC, 1999; Porstendörfer and Reineking, 1999; Nikezic et al., 2000; Yu et al., 2000b; Yu et al., 2001a; Yu et al., 2001b). Nonetheless, the ICRP commission — in its publication 65 (1994) — recommends the use of single conversion factor, derived mainly from epidemiological studies of uranium mine workers, for the

determination of the effective dose from inhalation of 222 Rn and 220 Rn progeny in private homes (members of the public) and workplaces (workers). Thus, a common assessment of the indoor effective dose, \mathbf{E}_{lung} (Sv), is given by the following equation

$$\mathbf{E}_{\text{lung}} = \text{DCF } \chi_{\text{D}} \tag{2.10}$$

where DCF is the dose conversion factors in units of Sv J⁻¹ m³ h⁻¹ for the potential α -energy exposure, $\chi_{\rm p}$, of indoor ²²²Rn and/or ²²⁰Rn daughter mixture.

Table 2.4 summarises the dose conversion factors for ²²²Rn and ²²⁰Rn decay products at private homes and workplaces according to the Spanish Legislation (BOE, 2001) that implements the council directive 96/29/EURATOM of the European Communities (OJEC, 1996)⁴. Outdoor exposure is generally neglected owing to its relative low levels of ²²²Rn, ²²⁰Rn and their progeny, and to the corresponding short occupancy time.

Table 2.4. Summary of the dose conversion factors for $^{222}\mathrm{Rn}$ and $^{220}\mathrm{Rn}$ progeny at private homes and workplaces.

	DCF (Sv $J^{-1} m^3 h^{-1}$)			
	222 Rn progeny	220 Rn progeny		
Workplaces	1.4	0.5		
Private Homes	1.1	0.4		

2.4. Emanation and transport within medium material

²²²Rn and ²²⁰Rn are originated from the radioactive decay of radium isotopes; themselves products of natural ²³⁸U and ²³²Th decay series in the earth's crust. Their indoor concentrations depend on the crustal abundance of their progenitors and on their access to building interiors. As a consequence, soils and rocks under or surrounding the dwellings are the main source of ²²²Rn and ²²⁰Rn to which people are exposed (Nazaroff et al., 1988a; UNSCEAR, 2000). The second sources of significance are building materials (Nazaroff et al., 1988a; Stranden, 1988; UNSCEAR, 2000). The contributions of other sources such as ground water, natural gases, and outdoor air are not ordinarily of relevant significance (Nero, 1988; Nazaroff et al., 1988b; UNSCEAR, 2000). Notwithstanding, there are circumstances in which these contributions are important and need to be considered.

Both soil and building materials can be divided into two major volume fractions: solid and porous fractions. The first fraction consists mainly of mineral grains of a wide

⁴The dose conversion factor for ²²⁰Rn daughters in private homes is estimated by dividing the corresponding workplaces' value by the ratio (1.4/1.1) of the DCF values given for ²²²Rn progeny.

range of sizes including a small amount of organic matter. The second fraction is usually occupied by water (moisture content) and gas, generally in similar composition to air. The porosity (η) , which is the fraction of the bulk volume of the considered soil matrix constituting the pore volume, increases with decreasing the solid grain size (Nazaroff et al., 1988a). A soil or a building material is saturated when the moisture content equals its porosity. Because soil and most earth-based materials have 10^3 - 10^4 times higher 222 Rn and/or 220 Rn concentration than the atmosphere, there is a substantial concentration gradient between such materials and indoor air. This gradient is permanently maintained by the generation of the 238 U and 232 Th series, from the long-lived parent radionuclides, and is responsible for a continuous flux of the radon isotopes. A schematic illustration of 222 Rn and 220 Rn emanation, transport and entry mechanisms from soil or buildings materials into indoor air is viewed in Figure 2.4. According to this Figure, the generation and migration of 222 Rn and/or 220 Rn into an indoor air depends on the following parameters:

- 1. The concentration of the immediate parent isotopes in the mineral grains 226 Ra for 222 Rn and 224 Ra for 220 Rn.
- 2. The extent to which the new formed atoms of ²²²Rn and ²²⁰Rn release from the solid mineral to the fluid-filled pores (emanation process).
- 3. The subsequent transport mechanisms through the pore volume.

2.4.1. Radium content

The radium content in soil and building materials is typically given as an activity per unit dry mass, $A_{\rm Ra}$ (Bq kg⁻¹). The most convenient and accurate method largely used for the determination of natural occurring radionuclides is by means of γ -spectrometry on individual samples previously dried, powdered and packed in small plastic containers with a given geometry (Nazaroff et al., 1988a; O'Connor et al., 1993; Ahmed et al., 1998; Ibrahim, 1999; Bojanowski et al., 2001). The presence of ²²⁶Ra in soil and building materials depends on their amount of ²³⁸U. At the same time, these two radioelements are often found to be in disequilibrium since they have different chemical properties, so that they must be considered separately. The ²²⁶Ra content is determined from the γ -emissions of the ²²²Rn progeny (²¹⁴Pb and ²¹⁴Bi) after being hermetically sealed and stored long enough to ensure equilibrium between ²²⁶Ra and ²²²Rn while the ²³⁸U is measured considering the γ -decay of its first short-lived daughter (i.e., the ²³⁴Th). Similarly, ²²⁴Ra can be assumed to be in secular equilibrium with ²²⁰Rn for sealed samples and its concentration can be obtained from the measurement of the ²²⁰Rn γ -emitter decay products (²¹²Pb, ²¹²Bi and ²¹²Tl). Nevertheless, the intermediate radioelements in the ²³²Th series to ²²⁴Ra have

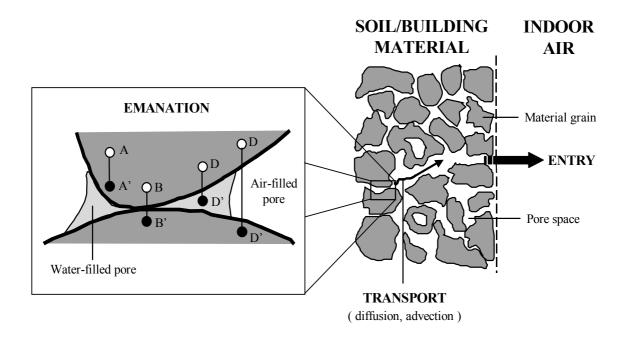


Figure 2.4. Schematic illustration of $^{222}\mathrm{Rn}$ and $^{220}\mathrm{Rn}$ emanation, transport and entry mechanisms from soil and buildings materials into indoor air — adapted from Knutson (1988). $^{226}\mathrm{Ra}$ ($^{224}\mathrm{Ra}$) atom, indicated by open circles, decays producing an α -particle and a $^{222}\mathrm{Rn}$ ($^{220}\mathrm{Rn}$) atom, which may end its recoil path at the point indicated by the solid circle. At A the parent atom is too deeply embedded within the grain for $^{222}\mathrm{Rn}$ or $^{220}\mathrm{Rn}$ atom to escape. At B and D the recoiling atom possesses sufficient energy after escaping the host to penetrate an neighbour grain. At C the $^{222}\mathrm{Rn}$ or $^{220}\mathrm{Rn}$ atom terminates its recoil in the pore water and, from there, it is readily transferred to the air-filled pore.

relatively short half-life in terms of geological time scales and are not altered to any extent by natural physical and/or chemical processes. Hence, a γ -spectrometry of the ²³²Th content considering the peaks of the intermediate γ -emitters, ²²⁸Ra and ²²⁸Ac, is a good indicator of the rate of ²²⁰Rn production within the reference soil or building material.

2.4.2. ²²²Rn and ²²⁰Rn emanation

Owing to the conservation of linear momentum, when radium isotopes decay within the material grains, the resulting atoms of ²²²Rn and ²²⁰Rn possess kinetic energies of 86 keV and 103 keV (see Appendix A), respectively. The newly formed atom travels from its site of generation until its recoil energy is transferred to the material. When the site of generation is close to the grain surface, a fraction of these atoms may reach the fluid-filled pore space (see Figure 2.4). This fraction is known as the *emanation coefficient* and is considered to have two components: recoil and diffusion. The recoil fraction itself can result in two fractions:

- i) a fraction of atoms that are stopped in the pore volume, and
- ii) another fraction where atom conserves a sufficient energy after escaping its host grain to traverse the pore space and to penetrate a neighbour grain.

The diffusion component refers to atoms that terminate their recoil in a solid grain — host or neighbour grain — and that may migrate towards the interstitial pore space by means of molecular diffusion. Nevertheless, as the diffusion coefficient of gases within the material grain is very low, this fraction is generally neglected. Therefore, the main component of the emanation coefficient is contributed mainly by the recoil fraction stopped in the pore volume. Similarly, any possible adsorption of ²²²Rn and/or ²²⁰Rn onto the soil grain surface can be neglected too — also because of their very low diffusion coefficient within the material grain.

The concentration of 222 Rn and 220 Rn in the air-filled pore volume increases with the soil moisture. This is due to the lower recoil range for 222 Rn and 220 Rn in water (\sim 723 Å and \sim 807 Å, respectively) than in air (\sim 65 μ m and \sim 76 μ m, respectively)⁵. A radon atom entering a pore that is filled or partially filled with water has a high probability of being stopped in the liquid phase without reaching another material grain. From there, it is readily transferred to the air-filled pore. The 222 Rn and 220 Rn equilibrium between the two phases, which decreases when increasing temperature, is achieved rapidly (Nazaroff et al., 1988a).

⁵The value of the recoil range of ²²²Rn and ²²⁰Rn in air and water were calculated using the Srim-2000 code based on the Bethe-Bloch formula given the stopping power of heavy charged particles in a medium — freely available in http://www.srim.org — (Ziegler and Biersak, 1985).

2.4.3. 222 Rn and 220 Rn transport

In order to enter the indoor air the ²²²Rn and ²²⁰Rn gas must be transported, basically through the larger air-filled pores within the soil or building material, so that a fraction of them reaches the soil-air or building-air interface before decaying. There are two basic mechanisms of ²²²Rn and ²²⁰Rn transport within medium material: diffusion and advection.

Transport by diffusion — Fick's law

The primary transport mechanism of 222 Rn and 220 Rn in a particular medium before decaying is done by the random molecular motion. Like any fluid substance there is a tendency to migrate in a direction opposite to that of the increasing concentration gradient within the material. This tendency is described by Fick's law, which relates the fluid flux density across the pore area to its concentration gradient. The coefficient relating these parameters is termed the effective diffusion coefficient, $D_{\rm e}$, with a proper unit of ${\rm m}^2 {\rm s}^{-1}$ and has an upper bound given by the diffusion coefficient in open air, $D_{\rm e}$.

According to the Fick's law, the diffusive flux density, $\overrightarrow{j_{\rm d}}$ (Bq m⁻² s⁻¹), of ²²²Rn or ²²⁰Rn activity per unit of pore area is

$$\overrightarrow{j_{\rm d}} = -D_{\rm e} \overrightarrow{\nabla} C \tag{2.11}$$

where C is the interstitial concentration of $^{222}\mathrm{Rn}$ or $^{220}\mathrm{Rn}$.

Many studies suggest that diffusion is the dominant mechanism by which radon enters the atmosphere from uncovered soil (Nazaroff et al., 1988a).

Transport by advection — Darcy's law

When a fluid has a sufficiently low Reynolds number, as is the case of $^{222}\mathrm{Rn}$ and $^{220}\mathrm{Rn}$ transport through soil and building material, viscous and/or laminar fluxes may be induced due to a pressure gradient. This gradient could be created mainly by changes in meteorological conditions and the use of mechanical systems such as exhaust fans or blowers, heating and air-conditioned systems in dwellings. Pressure-driven convective flow can be characterised by Darcy's law, which relates the apparent velocity of fluid flow through a cross-sectional area to the pressure gradient, $\overrightarrow{\nabla}P$, as follows

$$\overrightarrow{q} = -\frac{k}{\mu} \overrightarrow{\nabla} P \tag{2.12}$$

where \overrightarrow{q} (m s⁻¹) is the Darcy's velocity vector, which is defined as the flow per unit geometrical area over an element of volume large relative to individual pores; k (m²) is the

intrinsic gas-permeability; P is the pressure field in units of Pa; and μ (Pa s) is the dynamic viscosity of the gas. In this equation the effect of gravity is neglected.

The advective flow density per unit of pore area is then

$$\overrightarrow{j_{\rm a}} = C \overrightarrow{q} = -\frac{Ck}{\mu} \overrightarrow{\nabla} P \tag{2.13}$$

This expression is combined with equations of continuity and of state to obtain a governing formula for the pressure in the medium material. Considering the air-filled pore parts as incompressible, the pressure satisfies the following steady-state Laplace equation:

$$\overrightarrow{\nabla} \cdot \overrightarrow{q} = 0 \Rightarrow \nabla^2 P = 0 \tag{2.14}$$

The solution of this equation, for a specified geometry subject to appropriate conditions and assuming a constant pressure at the soil-air or building-air interface, is needed as input for the analysis of the general transport equation.

General transport equation

The concentration of 222 Rn and/or 220 Rn within a porous medium material is obtained from a general conservation-of-mass equation that combines the effects of diffusion and advection. Besides to affecting the 222 Rn and 220 Rn emanation process as described in Section 2.4.2, the presence of water may also alter the diffusion coefficient, the pressure field and the intrinsic permeability of the porous medium. In consequence, at large moisture content, the general transport equation must be corrected to account for these effects and for the partitioning of 222 Rn and/or 220 Rn between gas and liquid phases as follows (Nazaroff et al., 1988a)

$$\frac{1}{\eta} \frac{\partial (C_{\mathbf{a}} \eta_{\mathbf{a}} + C_{\mathbf{w}} \eta_{\mathbf{w}})}{\partial t} = D_{\mathbf{e}}' \nabla^{2} C_{\mathbf{a}} - \frac{k'}{\mu} \overrightarrow{\nabla} P \overrightarrow{\nabla} C_{\mathbf{a}} + \frac{1 - \eta}{\eta} \rho_{\mathbf{gr}} \lambda \widecheck{\mathbf{e}}' - \frac{\lambda}{\eta} (C_{\mathbf{a}} \eta_{\mathbf{a}} + C_{\mathbf{w}} \eta_{\mathbf{w}})$$
(2.15)

where: the primes indicate values accounting for the moisture content (Rogers and Nielson, 1991; Nazaroff et al., 1988a; Font, 1997),

 λ is the decay constant of ²²²Rn and/or ²²⁰Rn,

 $C_{\rm a}$ and $C_{\rm w}$ are the $^{222}{\rm Rn}$ and/or $^{220}{\rm Rn}$ concentration in the air-filled and water-filled pore parts, respectively,

 $\eta_{\rm a}$ ($\eta_{\rm w}$) is the air (water) porosity defined as the ratio of the air-filled (water-filled) pore volume to the bulk volume of the considered medium ($\eta = \eta_{\rm a} + \eta_{\rm w}$),

 $\rho_{\rm gr}$ (kg m⁻³) is the density of the mineral grains, and

ĕ (Bq kg⁻¹) is the emanation rate defined as the number of atoms that emanate into the pores per unit time and per unit mass.

Two implicit approximations were used in this equation. The first is that, as in the open air, all kinetic interactions of the radon atoms are assumed to occur with gas molecules. This is a reasonable approach since pore spaces in most soil and building materials are large relative to the mean free path of the radon atoms, which is comparable with that for the major constituents of air, $0.065~\mu m$ at Normal Temperature and Pressure (NTP)⁶. In the second simplification and taking into account the small diffusion coefficient in water than in air (see, for instance, Table 2.1), the diffusion motion of 222 Rn and 220 Rn atoms within the water-filled pore volume is neglected.

As an example, we consider the case of a dry and uncovered soil of semi-infinite depth. The steady-state one-dimensional solution of the Equation (2.15), assuming that radon migrates only by diffusion and that the concentration is equal to zero at the soil surface, is given by

$$C(x) = C_{\infty}(1 - e^{-\frac{x}{l_{\rm d}}}) \tag{2.16}$$

where x is the distance from the interface, , $C_{\infty} = \rho_{\rm gr} \frac{1-\eta}{\eta} \breve{\bf e}$ is the deep-soil $^{222}{\rm Rn}$ or $^{220}{\rm Rn}$ concentration being in secular equilibrium with radium and $l_{\rm d} = \sqrt{\frac{D_{\rm e}}{\lambda}}$ is the diffusion length. This last corresponds to the distance at which the deep-soil $^{222}{\rm Rn}$ or $^{220}{\rm Rn}$ concentration, C_{∞} , is reduced by a factor of $1-e^{-1}$. Taking as typical value for the effective diffusion coefficient, $D_{\rm e} = 2\times 10^{-6}~{\rm m}^2~{\rm s}^{-1}$ (UNSCEAR, 2000), the value obtained for $l_{\rm d}$ is $\sim 97.6~{\rm cm}$ for $^{222}{\rm Rn}$ and $\sim 1.3~{\rm cm}$ for $^{220}{\rm Rn}$. Consequently, in contrast to $^{222}{\rm Rn}$, only a small thickness of soil or building material serves as an effective source for $^{220}{\rm Rn}$.

By analogy, assuming a dominated pressure-driven flow with a constant Darcy's velocity and considering the same conditions as above, we can define the *advection length* as $l_{\rm a}=\frac{q}{\eta\lambda}$ (Font, 1997). In the case of being both diffusion and advection relevant transport mechanisms, the one-dimensional steady-state equation of $^{222}{\rm Rn}$ or $^{220}{\rm Rn}$ within a dry medium material — neglecting their concentrations at the soil surface — is

$$C(x) = C_{\infty} \left(1 - e^{-\frac{2x}{l_{\rm a} + \sqrt{l_{\rm a}^2 + 4l_{\rm d}^2}}} \right)$$
 (2.17)

⁶i.e., at 20 °C and 1 atm (Willeke and Baron, 1993; Friedlander, 2000).

and we define the migration length, $l_{\rm m}=\frac{1}{2}\left(l_{\rm a}+\sqrt{l_{\rm a}^2+4l_{\rm d}^2}\right)$, as a typical distance that $^{222}{\rm Rn}$ or $^{220}{\rm Rn}$ can migrate in the considered medium material and at which the deep-soil concentration is reduced by a factor of $1-e^{-1}$ (Font, 1997). Since the pressure gradient, the permeability and the effective diffusion coefficient may vary with time, we expect the migration length to have a high time-dependence.

2.5. ²²²Rn and ²²⁰Rn accumulation indoors

Once in indoor air and owing to their lack of chemical activity with other air components, ²²²Rn and ²²⁰Rn act as inert pollutants whose concentration is affected only by two factors: the entry and the removal processes. These factors are highly affected by the building design, the construction and operation characteristics, and the inhabitant behaviour as well.

2.5.1. Entry from soil

Figure 2.5. shows a schematic illustration of the 222 Rn and/or 220 Rn entry routes from soil into a house. We should note that the presence of a house may:

- i) alter the soil underneath changing its porosity,
- ii) serve as an umbrella to rain so that the soil moisture content is generally minimal, and
- iii) generate a small under-pressurisation (few Pascals) between indoor air and the soil underneath but high enough to provide all the radon needed to give the actual indoor levels.

Moreover, the building site itself may have graded or filled in a manner that the soil underlying characteristics are quite different from the original land patterns. Concrete is a very common material used in the building substructure, which acts as an effective barrier in front of ²²²Rn and/or ²²⁰Rn coming from the soil underneath taking into account its low diffusivity compared to that of soil (Rogers et al., 1995). The presence of cracks, gaps, holes and other penetration mechanisms in the slab may considerably increase the entry of ²²²Rn and/or ²²⁰Rn from the soil into an indoor air. Therefore, direct measurements of the resulting entry rate that depends strongly on the permeability of the underlying soil and on the under-structure characteristics of the building are not available. Consequently, a theoretical prediction of this source term for specific dwellings, taking into account all these effects and considering both diffusion and advection processes, is not easy to perform. The

possible contribution of 222 Rn and/or 220 Rn entry from soil can only be estimated from the observed total entry rate, subtracting the measured contribution of the building materials.

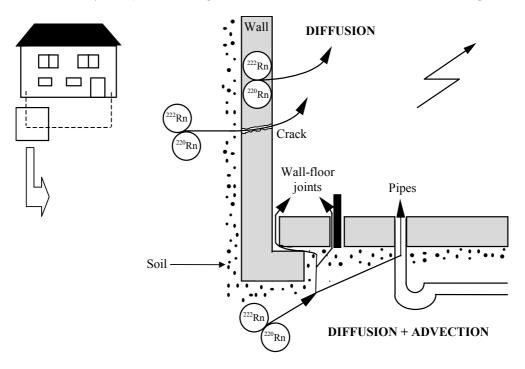


Figure 2.5. Schematic illustration of the ²²²Rn and/or ²²⁰Rn entry routes from soil into a house — adapted from (Font, 1997).

2.5.2. Entry from building materials

The rate of entry from building materials can be estimated from the exhalation rate $(e, \text{ in units of Bq m}^{-2} \text{ s}^{-1})$, which is defined as the activity released per units of geometrical surface and time. It is believed that the dominant transport mechanism of ^{222}Rn and/or ^{220}Rn within the building materials is by diffusion because most of these materials have very low permeability (Font, 1997). The water content in building materials is very small compared to that in soils. Neglecting the ^{222}Rn and/or ^{220}Rn concentration at both sides of the building material and the water content, the one-dimensional steady-state profile of ^{222}Rn and/or ^{220}Rn concentration is given by

$$C_{\rm B} = C_{\infty} \left[1 - \frac{\cosh\left(\frac{x}{l_{\rm d,B}}\right)}{\cosh\left(\frac{w_{1/2}}{l_{\rm d,B}}\right)} \right]$$
 (2.18)

where the subscript B stands for the building material, $w_{1/2}$ is its half-width and x is the distance from its centre. Accordingly, the exhalation rate obtained at both sides of the

building material is

$$e_{\rm B} = \eta_{\rm B} D_{\rm e,B} \left| \frac{\mathrm{d}C_{\rm B}}{\mathrm{d}x} \right|_{x=w_{1/2}} = \lambda \rho_{\rm gr,B} l_{\rm d,B} (1 - \eta_{\rm B}) \breve{\mathbf{e}}_{\rm B} \tanh \left(\frac{w_{1/2}}{l_{\rm d,B}} \right)$$
(2.19)

We should note that: i) the exhalation rate from building materials is affected by the type of surface coating or sealant used (Yu, 1993; Tso et al., 1994), and that ii) the exhalation rate of ²²⁰Rn from soils and building materials is generally 100 times greater than that of ²²²Rn (Porstendörfer, 1994).

2.5.3. Ventilation rate

Ventilation refers to the process whereby indoor air is replaced by outdoor air. It serves the purpose of controlling odors, maintaining a proper balance of metabolic gases and diluting pollutants generated by indoor sources. It is driven by pressure differences caused by the wind or by differences in the air temperature between indoors and outdoors. When the windows and doors are entirely open the indoor concentration is typically close to the outdoor levels. The ventilation rate, $\lambda_{\rm v}$ (h⁻¹), has a very important time-dependence and is made up of three components:

- 1. Infiltration and exfiltration of air by convection through small opening or imperfections (cracks) in the building materials.
- 2. Natural air exchange through windows or doors that are partially or temporarily opened.
- 3. Artificial ventilation supplied mechanically by the use of Heating, Ventilating and Air-Conditioned (HVAC) systems.

Characterising ventilation rate in dwellings is difficult since it is complex and has varied structure. The rates of air movement depend substantially on location, on meteorological conditions (wind speed, indoor-outdoor temperature difference), on the inhabitant activities, and on the HVAC systems used. However, if we assume that the outdoor air may enter and mix quickly so that it appears instantly throughout the volume of the given indoor space, the exact location of the leaks is not important. The time-averaged ventilation rate constant in realistic situations of occupied indoors, has been found to vary between 0.2 h⁻¹ to 2 h⁻¹ (ICRP, 1987; Knutson, 1988; Capra et al., 1994; Porstendörfer, 1994; UNSCEAR, 2000; Lembrechts et al., 2001). In artificially ventilated dwellings the ventilation rate can reach values about 10 times higher.

2.5.4. The indoor ²²²Rn and ²²⁰Rn concentration

As previously shown in this chapter, the indoor 222 Rn and/or 220 Rn concentration depends on the following parameters:

- 1. The entry rate of ²²²Rn and/or ²²⁰Rn from soil and building materials.
- 2. The exchange and dilution through mixing with the ambient air having much lower concentration.

In turn, these parameters are influenced by the characteristics (design, location, material of construction, occupant habits, etc.) of the dwellings chosen as a typical residential or working place. Since residences and workplaces are varied in structure and consist of a set interconnected rooms, the study of the spatial and temporal variation of the indoor ²²²Rn and ²²⁰Rn concentration is a complicated task. For simplicity, we shall consider only a single-room and, then, the real case of a multi-room building could be solved by taking into account the air exchange between the adjacent rooms (inter-zone flows). In this case, for each room the continuity condition must additionally be fulfilled.

Taking into account an uniform entry rate from soil and building materials, the time-behaviour of indoor 222 Rn or 220 Rn concentration is given by:

$$\frac{\partial C}{\partial t} = u_{\rm S} + \frac{e_{\rm B}S_{\rm B}}{V} - (\lambda + \lambda_{\rm v})C + \lambda_{\rm v}C^{\rm out}$$
(2.20)

where $u_{\rm S}$ is the volume-specific entry rate in Bq m⁻³ h⁻¹ of ²²²Rn or ²²⁰Rn in indoor air from soil, $S_{\rm B}$ is the surface area of the building materials, V is the air volume of the considered single-room and $C^{\rm out}$ is the outdoor concentration. This expression does not account for the fact that ventilation could influence the entry rates and assumes that there is not a pressure gradient within the considered air volume. Due to the relatively long half-life of ²²²Rn, an homogeneous activity distribution by complete mixing within the single room can be assumed. The only removal process it suffers in a non-ventilated room is the radioactive decay. However, the ²²²Rn radioactive decay constant ($\lambda \simeq 7.54 \times 10^{-3} \ h^{-1}$) is generally very small and can be ignored when compared to the common values of the ventilation rate. In the case of ²²⁰Rn ($\lambda \simeq 44.88 \ h^{-1}$), the removal effect by ventilation is very minimal.

In many cases, the indoor air movement and mixing is sufficiently fast and complete. The intensity or the degree of air mixing depends on the type of the HVAC systems used and on the indoor occupant activities. For that reason, there is no need to account for each crack and the exact entry point. The concentration of ²²²Rn and its progeny is normally homogeneously distributed within the considered air volume of the given room or

floor. The unique exception may be ²²⁰Rn which cannot be assumed to be uniformly distributed in all the indoor air volume owing to its short half-life (Doi et al., 1994; Yamasaki et al., 1995; Lida et al., 1996; Zhuo et al., 2001). Notwithstanding, since ²²⁰Rn decay products are longer lived, they would mix more or less homogeneously throughout the entire room volume and their concentration is assumed to be fractions of a representative average ²²⁰Rn concentration. The solution of the Equation (2.20) for indoor ²²²Rn and/or ²²⁰Rn concentration, assuming an instantaneous and homogeneous mixing of ²²²Rn and/or ²²⁰Rn within a reference room, has the following form

$$C(t) = \frac{u_{\rm S} + \frac{e_{\rm B}S_{\rm B}}{V} + \lambda_{\rm v}C^{\rm out}}{(\lambda + \lambda_{\rm v})} \left(1 - e^{-(\lambda + \lambda_{\rm v})t}\right)$$
(2.21)

Accordingly, under steady-state conditions, the ²²²Rn concentration is directly proportional to the entry rate from soil and building materials, decreasing reciprocally with the ventilation rate, while the ²²⁰Rn concentration is only affected by its radioactive decay. The time needed to establish the indoor concentration equilibrium within a non-ventilated room is of the order of several days in the case of ²²²Rn and several minutes in the case of ²²⁰Rn. On the other hand, in a ventilated room, the ²²²Rn reaches rapidly the equilibrium (the duration of which is of the order of several hours) while in the case of ²²⁰Rn the duration to the equilibrium does not change with respect to the non-ventilated room.

Table 2.5. Typical values and normal range of variation for the outdoor concentration, the soil volume-specific entry rate, the building material exhalation rate and the predicted indoor concentrations for ²²²Rn and ²²⁰Rn.

	$^{222}\mathrm{Rn}$		$^{220}\mathrm{Rn}$	
	Estimated	Range of	Estimated	Range of
	mean	variation	mean	variation
Outdoor Concentration (Bq m ⁻³)	1 - 10	1 - 20	1 - 5	1 - 10
$u_{\rm S} \; ({\rm Bq} \; {\rm m}^{-3} \; {\rm h}^{-1})$	1 - 40	1 - 500	_	_
$e_{\rm B} \; ({\rm Bq} \; {\rm m}^{-2} \; {\rm h}^{-1})$	1 - 5	1 - 20	20 - 120	10 - 1200
Indoor Concentration* (Bq m^{-3})	10 - 100	10 - 850	2 -10	1 - 100

^{*} Estimated using Equation (2.21).

Table 2.5 summarises typical values and normal range of variation for the outdoor concentration, the soil volume-specific entry rate, the building material exhalation rate and the predicted indoor concentrations for ²²²Rn and ²²⁰Rn (ICRP, 1987). The indoor concentrations were estimated by applying the steady-state approach of the Equation (2.21) and considering an average ventilation rate of 0.7 h⁻¹ to a reference room with 3 m long, 3

m wide and 3 m high. With these enclosure dimensions the indoor air volume is $V=27~\mathrm{m}^3$ surrounded by a surface area of $S_\mathrm{B}=54~\mathrm{m}^2$. Because of its short radioactive half-life, the entry rate of $^{220}\mathrm{Rn}$ from soil is neglected. However, in dwellings with no efficient barriers between soil and indoor air space, it cannot be excluded as the rate of its entry rate might be comparable with that of $^{222}\mathrm{Rn}$.