3. Properties and behaviour of the airborne ^{222}Rn and ^{220}Rn progeny

The nature and behaviour of the airborne ^{222}Rn and ^{220}Rn decay products are much more complex in comparison with their noble gas parents and are governed by physical and/or chemical interactions with air constituents. At their birth, they may participate in one or more of the following processes — not necessarily in the order stated: i) neutralisation, ii) nucleation or formation of the clusters, iii) attachment (detachment) to (from) airborne aerosol particles, iv) deposition on any available surface in direct contact to indoor air (floor, ceilings, walls and area of other objects such as furniture), and v) radioactive decay. Furthermise, they may be transported to the oudoor air by convective flows due mainly to ventilation.

The process of radioactive aerosol formation is considered to be a special case of airborne particle coagulation (Willeke and Baron, 1993; Friedlander, 2000). Following aerosol-attachment, the behaviour of the ^{222}Rn and ^{220}Rn progeny depends on the carrier aerosol particle characteristics and can be treated as being submersed in a continuous gaseous medium or, more broadly, a fluid (the continuum regime). Because of their small size and mass, of the same order of the gas molecules, the airborne-unattached ^{222}Rn and ^{220}Rn progeny have higher diffusitivities than the aerosol-attached decay products and diffuse more readily to surfaces.

3.1. Airborne-unattached ^{222}Rn and ^{220}Rn daughters

By unattached or free decay products we refer to airborne ^{222}Rn and/or ^{220}Rn progeny charged or neutral, in any physico-chemical state, that are not attached to aerosol particles. According to this definition, progeny subject only to chemical reaction (ultrafine mode) or to molecular clustering (cluster mode) in the air would be defined as airborne-

unattached. As the behaviour of airborne ^{222}Rn and ^{220}Rn progeny is very complex, its main feature may be illustrated by considering only the first short-lived daughter in the ^{222}Rn chain (i.e., the ^{218}Po). The same argument found for this isotope can be applied to all the remaining decay products.

As shown in Chapter 2, ^{222}Rn atom decays with the emission of an α-particle having an energy of 5.49 MeV. The resulting ^{218}Po atom has a recoil energy of 101 keV (see Appendix A) sufficient to produce a recoil range[1] of 72 μm in air or of 0.04 μm in a particle of density 2 g cm^{-3}. The freshly generated radionuclide interacts with the air molecules during its recoil path (the duration of which is of the order of nanoseconds), loses its charge and tends to be positively ionised with a single positive charge after thermalisation (Raabe, 1969). The charged fraction of this radionuclide was found to decrease with the increasing Relative Humidity (RH) and is assumed to be equal to $\sim$90% at Normal Temperature and Pressure (NTP) (Phillips et al., 1988).

3.1.1. Neutralisation of positively charged ^{222}Rn and ^{220}Rn progeny

Neutralisation of positively charged ^{222}Rn and ^{220}Rn progeny in air is a complicated process not well understood yet. It may involve one or more mechanisms depending on the ionisation potential of the ^{218}Po species and of the gas molecules, on the concentration of its parent (^{222}Rn) in the gaseous environment, on the air relative humidity and on the concentration and nature of the airborne negative ions. The three mechanisms commonly proposed to describe the ^{218}Po^{+} neutralisation are (Frey et al., 1981; Busigin et al., 1981; Chu and Hopke, 1988):

1. recombination with airborne small negative ions,

2. electron-scavenging by means of the airborne OH radicals, and

3. charge transfer with airborne neutral atoms.

Recombination with airborne small negative ions

In this mechanism, the airborne ^{218}Po ions may recombine with small negative ions (including electrons). These last are produced as a result of the air ionisation from α-emission as well as the ^{222}Rn and/or ^{220}Rn daughter recoil process. The airborne small negative ion concentration is given by (Busigin et al., 1981)

$$\frac{\partial n_-}{\partial t} = \xi n_0 - \alpha n_+ n_- \tag{3.1}$$

---

[1]The recoil ranges were calculated using the Srim-2000 code.

where ξ is the ion pair production rate as a result of α-decay of ^{222}Rn or ^{220}Rn,

n_0 is the ^{222}Rn or ^{220}Rn concentration in units of cm^{-3},

α is the ion pair recombination coefficient, $\alpha = 1.4 \times 10^{-6}$ cm^3 s^{-1} according to Chu and Hopke (1988), and

n_+ and n_- are respectively the concentration of positive and small negative ions in air.

To give an example of illustration of this effect, we take the case of airborne ion pair production as a result of ^{222}Rn decays. Subsequently, neglecting the energy loss due to elastic collisions with the airborne molecules and taking into account that the air average ionisation potential is $\sim$34 eV, each ^{222}Rn α-emission produces approximately 2×10^5 of airborne ion pairs resulting in an ion pair production rate of $\sim$0.42 s^{-1} per atom of ^{222}Rn.

Assuming the same airborne concentration for both positive and small negative ions, $n_+ \simeq n_-$, the steady-state solution of Equation (3.1) yields

$$n_+ = n_- = \sqrt{\frac{\xi n_0}{\alpha}} \tag{3.2}$$

The neutralisation rate, K, is proportional to the airborne negative ion concentration present in air and therefore

$$K \propto \sqrt{\frac{\xi n_0}{\alpha}} \tag{3.3}$$

where the proportionality constant has been estimated to be of the order of 10^{-5} (Busigin et al., 1981; Chu and Hopke, 1988; Shi and Hopke, 1991) suggesting that the neutralisation mechanism with airborne small negative ions may be significant only at high concentration of ^{222}Rn or ^{220}Rn atoms (of the order of kBq m^{-3}). It has been also shown that this mechanism dominates only at relative humidities above 20% (Chu and Hopke, 1988; Shi and Hopke, 1991). Nevertheless, at lower humidities, neutralisation of ^{218}Po$^+$ still not obeying Equation (3.3) and, in turn, has a high dependence on the air humidity (George and Breslin, 1977). This fact has let to suppose the existence of other neutralisation mechanisms such as electron-scavenging by means of the airborne OH radicals, and charge transfer with airborne neutral atoms.
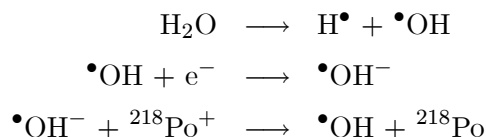
Electron-scavenging by means of the airborne OH radicals[2]

Frey et al. (1981) suggested that neutralisation may also occur as a result of electron-scavenging by means of airborne water vapour molecules. Nonetheless, Goldstein

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[2]The term *radical* or *free radical* is used for a neutral atom or molecule in which one of the bonding orbitals is occupied by a single electron.

and Hopke (1985), pointed out that it cannot be the water vapour molecule itself that is trapping the electrons in air. Radiolysis of the water and/or alcohol molecules leads to formation of the hydroxyl (OH) radicals, which are good electron acceptors (with an electron affinity of 1.83 eV). The airborne free electrons, as shown above, are originated from the gas ionisation due to the parent (^{222}Rn) decay. Accordingly, the formation of airborne OH radicals plays an important role both in the neutralisation process for the polonium ion and in the reaction chemistry leading to the formation of radiolytic components. In the presence of water vapour molecules only, the mechanism of electron scavenging can be explained according to the following chemical reactions (Chu and Hopke, 1988):

$$
\begin{aligned}
\mathrm{H_2O} &\longrightarrow \mathrm{H^{\bullet}} + \mathrm{^{\bullet}OH} \\
\mathrm{^{\bullet}OH} + \mathrm{e^-} &\longrightarrow \mathrm{^{\bullet}OH^-} \\
\mathrm{^{\bullet}OH^-} + \mathrm{^{218}Po^+} &\longrightarrow \mathrm{^{\bullet}OH} + \mathrm{^{218}Po}
\end{aligned}
$$

Another way to validate this hypothesis is by adding the alcohol (ethanol) volatile molecules able to react with $^{\bullet}$OH group of the above reaction. By the introduction of alcohol volatile molecules, the last two reactions may be dominated by the preferred reaction

$$
\mathrm{ROH} + \mathrm{^{\bullet}OH} \longrightarrow \mathrm{RO^{\bullet}} + \mathrm{H_2O}
$$

and the neutralisation rate was then reduced to value ranges similar to those obtained in the small negative-ion recombination mechanism (Chu and Hopke, 1988).

The OH radical concentration can be evaluated from the equation

$$
\frac{\partial [\mathrm{^{\bullet}OH}]}{\partial t} = Q[\mathrm{H_2O}] - \mathrm{k_R}[\mathrm{H^{\bullet}}][\mathrm{^{\bullet}OH}] \tag{3.4}
$$

where Q is the dissociation constant of the water vapour molecules and $\mathrm{k_R}$ is the recombination constant of the radicals $\mathrm{H^{\bullet}}$ and $^{\bullet}$OH with airborne concentrations $[\mathrm{H^{\bullet}}]$ and $[^{\bullet}\mathrm{OH}]$, respectively.

Assuming the same concentration level for the radicals $\mathrm{H^{\bullet}}$ and $^{\bullet}$OH, i.e., $[\mathrm{H^{\bullet}}] = [^{\bullet}\mathrm{OH}]$, the steady-state solution of Equation (3.4) gives

$$
[\mathrm{^{\bullet}OH}] = \sqrt{\frac{Q[\mathrm{H_2O}]}{\mathrm{k_R}}} \tag{3.5}
$$

which suggests that the $^{\bullet}$OH radical concentration in air and, hence, the neutralisation rate should be proportional to the square root of the water vapour concentration. The same behaviour was found in dry nitrogen (Chu and Hopke, 1988). As $\mathrm{O_2}$ and $\mathrm{NO_2}$ are

known to be excellent electron acceptors — with an electron affinity of 0.44 eV and 2.32 eV, respectively — we can conclude that electron-scavenging by means of these elements is feasible also.

Charge transfer with airborne neutral atoms

Another mechanism of ^{218}Po neutralisation in air, is by removing electrons directly from colliding neutral molecules. However, this mechanism is unlikely since the first ionisation potential of polonium ($\sim$8.43 eV) is lower than that of various airborne molecules. The only exceptions are the hydrocarbon air components, which have even lower ionisation potentials. Instead, Busigin et al. (1981) suggested that, being the ^{218}Po$^+$ unstable, it may react chemically with the air oxygen to form polonium dioxide (PoO_2^+) for which they estimated an ionisation potential of $\sim$10 eV. In such circumstances, the polonium dioxide can become neutralised by removing electrons from trace gases having a substantially lower ionisation potential (such as NO, NO_2, NH_3 or SO_2) and/or water vapour. Other chemical reactions with atmospheric constituents are possible. In the presence of carbon monoxide, polonium carbonyl may form with ionisation potential high enough.

Even if the charge transfer with airborne neutral atoms is the dominant neutralisation process (Chu and Hopke, 1988), it depends mainly on the air ionisation potential, on its content of trace gases and/or organic vapours, and on the concentrations of the airborne ^{222}Rn and/or ^{220}Rn. This neutralisation is faster since the electrons can be transferred directly by a single collision with a donor molecule. The time needed for the polonium neutralisation in air is of the order of milliseconds (Phillips et al., 1988).

3.1.2.   Cluster formation (nucleation)

Nucleation or nucleated condensation refers to the process of initial formation of a particle from the gas phase. Evaporation is the reverse process of condensation, and results in a net loss of molecules and a shrinkage of the particle. The airborne ^{218}Po$^+$ — as well as the PoO_2^+ — that serves as a site for condensation may grow spontaneously by the clustering of polar molecules around it to form stable complexes. This process, in which water vapour (H_2O) have an important participation, may be very compatible with all the neutralisation mechanisms discussed above. The duration of the cluster formation is estimated to be between 5×10^{-6} s and 5×10^{-7} s (Raes et al., 1985); so that it can be considered to take place mainly before the neutralisation. When the cluster is neutralised, it will evaporate and will result in a neutral atom.

Even if the nucleation process is discret, its interpretation is usually carried out using the classical thermodynamic theory of liquid drop formation. In the following, we

present this theory, in simplistic terms, to show the parameters influencing the nucleation process. The Gibbs free energy of a pure H_2O cluster formation with a radius r is (Bricard, 1977; Friedlander, 2000)

$$\Delta G = -\frac{4\pi r^3}{3m_{H_2O}}\rho_{clst}\kappa T \ln \frac{P}{P_s} + 4\pi r^2 \gamma_s + \frac{q^2}{2}\left(\frac{1}{\epsilon_0} - \frac{1}{\epsilon}\right)\left(\frac{1}{r} - \frac{1}{r_0}\right) \qquad (3.6)$$

where m_{H_2O} is the molecular weight of water, ρ_{clst} is the cluster density, T is the absolute temperature of the air, P is the ambient water vapour pressure, P_s is the vapour pressure on a flat surface of water at which the evaporation and the condensation reach an equilibrium ($\frac{P}{P_s}$ is the saturation degree), κ is the Boltzmann constant, γ_s is the cluster superficial tension, q is the cluster charge, ϵ_0 and ϵ are the dielectric constants of the vapour and droplet substance, respectively, and r_0 is the ion radius.

The first term at the right of Equation (3.6) takes into account the liberation energy of the condensation by latent heating. The second term calculates the surface free energy of a liquid drop with a radius r. The third and the last term refers only to the ion nucleation process and describes the electrical energy change owing to the presence of water molecules. According to this equation and to the results found by Raes et al. (1985) — under unsaturated conditions (below 100% RH) — the formation of neutral H_2O clusters in air is thermodynamically impossible. The ionised clusters grow until they reach an equilibrium size at which the number of molecules evaporating from the cluster is equal to the number of molecules that condense on its surface. The cluster radius, which is very influenced by the air humidity, can be obtained by resolving the following expression

$$\frac{\partial \Delta G}{\partial r} = 0 \qquad (3.7)$$

Under normal conditions, the cluster diameters ($d = 2r$) are estimated to be between 1.3 nm for 1% RH (40 water molecules forming the cluster) and 2.3 nm for 100% RH (220 water molecules in the cluster) (Phillips et al., 1988).

Another possibility of ^{222}Rn and/or ^{220}Rn progeny clustering is by the condensation of other airborne species such as CO_2, H_2SO_4, HNO_3 and organic vapours. Notwithstanding, in contrast with a pure H_2O cluster, there is no evaporation after neutralisation; so that neutral cluster nucleation is feasible. The neutralised cluster will continue to grow forming a drop aerosol.

3.1.3. Diffusitivity of the airborne-unattached ^{222}Rn and ^{220}Rn progeny

As the unattached fraction of the airborne ^{218}Po has molecular size (below 10 nm) under realistic environmental conditions (Porstendörfer, 1994; Mohammed, 1999; Tymen et

al., 1999; Cheng et al.,2000; Huet et al., 2001), the ^{218}Po atoms are affected by the motion of individual gas molecules (the free molecular regime). Afterwards, the kinetic theory of gases is useful in understanding the behaviour of these particles. The diffusion coefficient, D, of any particle suspended in a gas medium is commonly estimated by the Stokes-Einstein equation

$$D = \frac{\kappa T \mathbf{CU}}{3\pi\mu d_{\mathrm{p}}} \tag{3.8}$$

where μ is the gas viscosity (1.83×10^{-4} g cm s^{-1} for air), d_{p} is the particle diameter and $\mathbf{CU}$ is the *Cunningham* slip correction factor, which is related to the particle diameter (Allen and Raabe, 1985; Cheng et al., 1988), as

$$\mathbf{CU} = 1 + \frac{2l}{d_{\mathrm{p}}}\left(1.142 + 0.558 \ e^{-0.999\frac{d_{\mathrm{p}}}{2l}}\right) \tag{3.9}$$

where l is the mean free path of the gas, i.e., the mean travelling distance of a gas molecule before colliding with another molecule — $\sim$0.065 μm for the air at NTP (Willeke and Baron, 1993; Friedlander, 2000).

Equation (3.9) was empirically derived to fit the entire range of $\frac{d_{\mathrm{p}}}{2l}$ values, i.e., from the continuum to the free molecular regimes. However, Equations (3.8) and (3.9) overestimate the particle diffusion coefficient of molecular clusters having diameter smaller than 2 nm. To correct this overestimation, d_{p} may be substituted in Equation (3.9) for the Cunningham constant by d_{p}^{*} having the following expression (Ramamurthi and Hopke, 1989)

$$d_{\mathrm{p}}^{*} = d_{\mathrm{p}}\left(1 + 3 \ e^{-2.2\times10^{7}d_{\mathrm{p}}}\right) \tag{3.10}$$

Since 1956, a number of studies concerning the diffusion coefficient of the airborne-unattached ^{218}Po progeny has been performed with partly contradictory results. Most of these studies are summarised by Phillips et al. (1988) and the measured values obtained range from 0.01 cm^2 s^{-1} to 0.1 cm^2 s^{-1}, depending on the experimental conditions with regard to the chemical and physical state of the air. During the last decade, the scarce measurements performed for the diffusion coefficient of airborne-unattached ^{222}Rn and ^{220}Rn decay products confirm the validity of this range of variation (George and Knutson, 1994; Leonard, 1996; Knutson et al., 1997; Tokonami, 1999; Tymen et al., 1999; Malet et al., 2000). Furthermore, the presence of charge on the daughter atom increases its interaction with surrounding air ions resulting in a lower diffusivity of this atom in the gas medium. For that reason, the diffusion coefficient of a charged airborne-unattached ^{222}Rn and/or ^{220}Rn decay products is smaller than that of a similar atom with a neutral charge.

It may be concluded that there is no single diffusion coefficient for the ^{222}Rn and/or ^{220}Rn progeny. Both the electric charge and size of the unattached ^{222}Rn and/or ^{220}Rn decay products determine their diffusion coefficient in air. Each value of this diffusion coefficient must be regarded as a particular case for a given set of chemical and/or physical conditions. The use of a single value may therefore be used with caution if we want to represent realistic situations of indoor environment.

3.2. Formation of ^{222}Rn and ^{220}Rn daughter aerosols

3.2.1. The indoor aerosol particles

Besides the fast reactions of neutralisation and cluster formation, the decay products of ^{222}Rn and ^{220}Rn participate in the normal kinematic behaviour of the air and attach to existing particles of the airborne aerosol. The term *aerosol* — from the Greek word combination air-particle — refers to an assembly of liquid or solid particles suspended in the air. Aerosol particles are generally originated from natural and man-made sources in both outdoor and indoor environments. Those found in outdoor or ambient air stem largely from soil dusts, sea salts, combustion products, gas emissions from stationary and mobile sources such as power plants and cars, forest fires, volatile substances and biological materials (e.g., pollen, spores, bacteria, etc.) (Willeke and Baron, 1993; Maroni, 1998; Friedlander, 2000).

The indoor aerosol particle concentration depends mainly on the outdoor particle concentration, on the ventilation rate and on other indoor sources owing to the occupant habits. Cooking, smoking and the use of spray products are the prominent source of indoor aerosols. Since aerosols are extremely varied in shape, it is common to assign an equivalent diameter which is reported as the diameter of a sphere having the same value of a specific physical property as that one can measure or calculate. For example, when the random molecular motion is of concern, the diffusion-equivalent diameter is the diameter of a unit-density sphere with the same rate of diffusion as the particle in question (Willeke and Baron, 1993). The aerosol particles range in size from almost molecular dimensions up to several micrometers in diameter (Willeke and Baron, 1993; Dorrian, 1997; Friedlander, 2000).

3.2.2. Attachment to aerosol particles

Aerosol-attachment is the process whereby an airborne-unattached ^{222}Rn and ^{220}Rn daughter atom or cluster, which undergoes random motion like any gas molecule, strikes and sticks to an aerosol particle. To date, the most authoritative version of the aerosol-attachment theory seems to be that presented by Porstendörfer (1994). In this theory, latter verified experimentally by Tokonami (2000) at the Environmental Measurement

Laboratory (EML, USA) radon/aerosol facility, the sticking probability of airborne decay products on the aerosol surface is taken as unity. Given $\Delta Z(d_p) = Z(d_p)\Delta(d_p)$ the aerosol particle concentration — in units of cm^{-3} — in the size interval Δd_p of the particle diameter d_p and number concentration $Z(d_p)$, the aerosol-attachment rate, $\lambda_a(d_p)$ (h^{-1}), that expresses the adsorption velocity of the unattached decay products to aerosol particle in air is given by

$$\lambda_a(d_p) = \beta(d_p)\Delta Z(d_p) = \beta(d_p)Z(d_p)\Delta(d_p) \tag{3.11}$$

where $\beta(d_p)$ (cm^3 h^{-1}), which is a function of particle aerosol size d_p, is known as the aerosol-attachment coefficient. The theory of aerosol-attachment is based on random collisions that occur between the airborne-unattached ^{222}Rn and/or ^{220}Rn progeny, aerosol particles, and gas molecules. Hence, the aerosol-attachment coefficient is satisfactorily described by the following expression

$$\beta(d_p) = \frac{\pi D v_0 d_p^2 (\frac{d_p}{2} + l)}{4D(\frac{d_p}{2} + l) + \frac{v_0 d_p^2}{4}} \tag{3.12}$$

where v_0 is the mean thermal velocity of the airborne-unattached ^{222}Rn and ^{220}Rn progeny (1.72×10^4 cm s^{-1} at NTP) (Porstendörfer, 1994). There are two limiting values to equation (3.12):

(1) For small particles ($d_p \ll l$), the aerosol-attachment is controlled by pure kinetic theory of gases and is proportional to the aerosol surface:

$$\beta(d_p) = \frac{1}{4}\pi v_0 d_p^2 \tag{3.13}$$

(2) For large particles ($d_p \gg l$), the aerosol-attachment process is determined by pure diffusion laws:

$$\beta(d_p) = 2\pi D d_p \tag{3.14}$$

Usual assumptions taken in the above theories are that i) the size and mass of the airborne-unattached ^{222}Rn and ^{220}Rn daughters are negligibly small compared to those of an aerosol particle, ii) the electrostatic forces need not be taken into account, and iii) the airborne-unattached decay products concentration is low enough that the corresponding agglomeration can be neglected.

A single value of the aerosol-attachment coefficient is, therefore, valid only for monodisperse aerosols. In practice, real aerosol particles in home, buildings and workplaces are polydisperse. As a consequence, the total number particle concentration is obtained by

integration over the whole size distribution of the indoor aerosols ($Z = \int_{d_\mathrm{p}} Z(d_\mathrm{p})\,\mathrm{d}d_\mathrm{p}$) and the corresponding aerosol-attachment rate can be written as

$$\lambda_\mathrm{a} = \int_{d_\mathrm{p}} \beta(d_\mathrm{p}) Z(d_\mathrm{p})\,\mathrm{d}d_\mathrm{p} \qquad (3.15)$$

To better represent the actual polydisperse aerosols of indoor air, the determination of an average aerosol-attachment coefficient, β, is required. Taking a typical value of $\beta = 5 \times 10^{-3}$ cm^3 h^{-1} (Porstendörfer, 1994), the aerosol-attachment rate might range from as low as 5 h^{-1} (at $Z = 10^3$ cm^{-3}) in clean air to 500 h^{-1} at high aerosol concentration ($Z = 10^5$ cm^{-3}). The latter case of high indoor aerosol concentration arise mainly because of cooking or cigarette smoking activities.

3.2.3. Desorption from aerosol particles (recoil factor)

The α-recoil factor, p, defines the probability of an aerosol-attached ^{222}Rn and ^{220}Rn daughter to desorb from its host as a consequence of an α-decay of its precursor. As previously shown in this chapter, the α-recoil energies are between 101 keV and 128 keV in ^{222}Rn and ^{220}Rn decay series (see Appendix A). These energies are high compared to the potential energy of a molecule absorbed on a surface (of the order of eV). Thus, in the case of α-emissions from airborne ^{222}Rn and/or ^{220}Rn progeny, which are attached to aerosol, a certain fraction of the resulting atom may become detached from its carrier aerosol.

The α-recoil mechanism has been investigated experimentally suggesting that it depends on the nuclear stopping power in aerosol and on particle size (Mercer, 1976; Kojima, 1988). According to these experiments, the α-recoil fraction from aerosol particles is taken to be 0.8. Upon decay of aerosol-attached ^{222}Rn and/or ^{220}Rn progeny by means of β-emissions, the resulting atoms may also become unattached on recoil. Nevertheless, their recoil is much smaller than that of α-emissions and no experimental evidence has been found to support the aerosol detachment after β-decay. The β-recoil fraction is therefore taken to be zero.

3.3. Deposition on surfaces

Airborne ^{222}Rn and ^{220}Rn decay products, whether unattached or attached to aerosol particles, tend to deposit on any available surface in direct contact to indoor air, becoming no longer available for inhalation and causing lung doses. The term *plate-out* is often used for surface deposition process of airborne ^{222}Rn and/or ^{220}Rn progeny. Following plate-out, re-entrainment into the air owing to α-recoil mechanism is neglected in most cases. Assuming always well-mixed indoor air and that all the surface areas in the reference room

are equally effective in collecting ^{222}Rn and/or ^{220}Rn decay products, the respectif loss rates by plate-out effect of these nuclides in airborne-unattached and aerosol-attached state are, respectively, given by

$$\lambda_{\mathrm{d}}^{\mathrm{u}} = v_{\mathrm{d}}^{\mathrm{u}}\frac{S}{V} \text{ and } \lambda_{\mathrm{d}}^{\mathrm{a}} = v_{\mathrm{d}}^{\mathrm{a}}\frac{S}{V} \qquad (3.16)$$

where the superscripts u and a refer to airborne-unattached and aerosol-attached state of the considered daughter atom, respectively, S includes the area of the walls, floor, ceiling and furniture in the reference room available for deposition, V is the indoor air volume and $v_{\mathrm{d}}^{\mathrm{u}}$ ($v_{\mathrm{d}}^{\mathrm{a}}$) is the deposition velocity of airborne-unattached (aerosol-attached) ^{222}Rn and/or ^{220}Rn progeny.

Due to aerosol large sizes (between 0.1 μm and 10 μm) (Tymen et al., 1992; Porstendörfer, 1994; Dorrian, 1997; Tokonami et al., 1997; Harley et al., 2000; Shimo and Saito, 2000) and consequently low diffusion coefficients (from 2×10^{-8} cm^2 s^{-1} to 7×10^{-6} cm^2 s^{-1}) (Willeke and Baron, 1993; Friedlander, 2000), deposition velocities for aerosol-attached progeny are much smaller than those for the airborne-unattached species (approximately two orders of magnitude smaller). In addition, the plate-out effect may be enhanced as a result of indoor air flow patterns, or of the presence of fortuitous electric fields due to domestic electrical appliances and/or to electrostatic charge on the available surfaces. As a consequence, it is difficult to predict with accuracy the expected values of the deposition velocities in all indoor environments. This difficulty also arises because very few direct measurements of plate-out velocities in real situations have been made. In turn, since 1980 efforts have been invested to obtain experimental values in small exposure chambers where it is practical to exclude aerosols; so that aerosol-attachment has not to be taken into account (Bruno, 1983; Knutson et al., 1983; Gadgil et al., 1992; McLaughlin and Fitzgerald, 1992; Kojima et al., 1993; Nazaroff et al., 1992; Nazaroff et al., 1993; Hattori and Ishida, 1994; Porstendörfer, 1994; Xu et al., 1994; Thatcher et al., 1996; Leonard, 1996; Voytchev et al., 1999; Schmidt and Hamel, 2001). The reported values suggest quite strongly that the deposition velocity of the unattached ^{222}Rn and/or ^{220}Rn daughters in air, is in the range of 0.05 - 0.5 cm s^{-1}. For aerosol-attached progeny, the reported data of the deposition velocity are less consistent with possible values ranging from 5×10^{-4} to 5×10^{-3} cm s^{-1}.

The surface-to-volume ratio, $\frac{S}{V}$, depends on the size and on the geometry of the reference room as well as on the furniture used within this last. For a sphere or cube with a diameter or side-length a we have $\frac{S}{V} = \frac{6}{a}$. The latter geometry might be considered in most privates homes and workplaces. Taking into account that any furniture used may increase the indoor surface area and may decrease the air volume, a larger surface-to-volume ratio of $\frac{S}{V}$ between 3 m^{-1} and 6 m^{-1} has to be considered in most typical dwellings (Porstendörfer,

1994). With these values and the reported plate-out velocities given above, the deposition rate for the airborne-unattached and aerosol-attached ^{222}Rn and/or ^{220}Rn decay products may range from 5 h^{-1} to 110 h^{-1}, and from 0.05 h^{-1} to 1.1 h^{-1}, respectively.

3.4. Removal by ventilation

As discussed in Chapter 2, ventilation is the process whereby indoor air is replaced by outdoor air. It is also an important parameter for understanding the mixing and dilution of the indoor ^{222}Rn and ^{220}Rn progeny concentration. Like their parent atoms, the airborne ^{222}Rn decay products can leave indoor air by exfiltration through cracks on the building structure, by natural air exchange through windows or doors, and/or by artificial exhaust fans or blowers. It is generally assumed that the daughters exhausted by the ventilation do not return indoors. In the case of the airborne ^{220}Rn progeny, although the ventilation effect is minimal for the noble gas parent, it is useful for reducing their concentration, especially for the ^{212}Pb whose half-life is longer compared with other daughters. However, for airborne-unattached ^{222}Rn and ^{220}Rn decay products, the removal effect by ventilation can be ignored with respect to the common value of their deposition rate. Otherwise, it has been indicated that the ventilation process may increase the diffusion coefficient of airborne-unattached ^{222}Rn and ^{220}Rn daughters (Kotrappa et al., 1976; Bruno, 1983). Consequently, the deposition velocity of these decay products may increase as well.

3.5. The indoor ^{222}Rn and ^{220}Rn daughter concentration

Alongside this chapter, it has been made evident that — in contrast to ^{222}Rn and ^{220}Rn, which are not influenced by the air constitution and characteristics — the nature and behaviour of their airborne progeny is further complicated due mainly to the diverse interactions that they experiment with the surrounding environment. Figure 3.1 illustrates the fundamental processes influencing the indoor concentrations of the airborne ^{222}Rn and ^{220}Rn progeny. It can be clearly seen, in this figure, that the ^{222}Rn and/or ^{220}Rn progeny being in airborne-unattached form may participate in the following processes:

- to attach to any neighbouring aerosol particles,

- to deposit on the available surfaces (walls, floor, ceiling, and furniture),

- to be exhaust throughout the outdoor air by means of ventilation, and

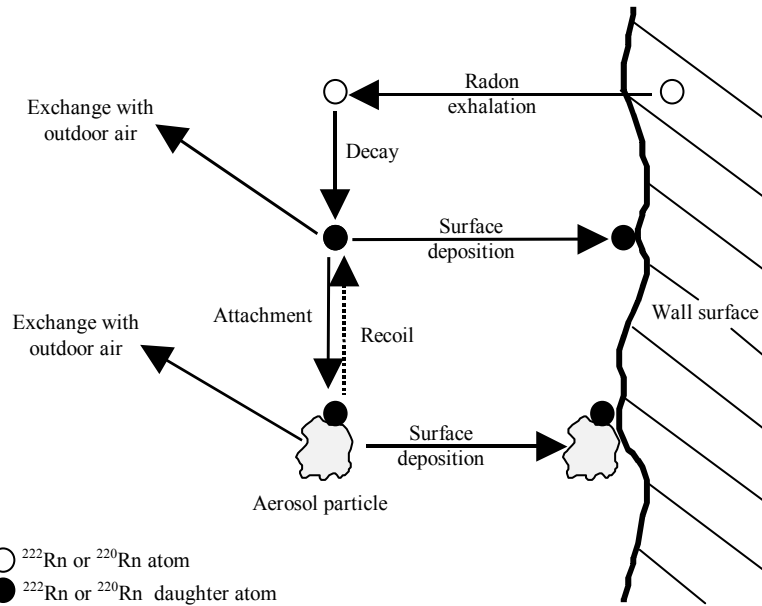- to decay to the following daughter.

Figure 3.1. The basic processes influencing the indoor activity balance of ^{222}Rn and ^{220}Rn progeny — adapted from Knutson (1988).

On the other hand, the ^{222}Rn and/or ^{220}Rn decay products attaching to aerosol particles may have the following alternatives:

- to detach from their host aerosol because to the recoil energy acquired along the decay of their precursor, especially in the case of α-emissions.

- to deposit on the available surfaces (walls, floor, ceiling, and furniture),

- to be exhaust throughout the outdoor air by means of ventilation, and

- to decay to the following daughter.

A theoretical approach, which is widely used to predict the indoor ^{222}Rn and ^{220}Rn progeny behaviour, is that developed by Jacobi (1972). In this approach, the time dependence of the airborne ^{222}Rn and ^{220}Rn daughter concentration within a reference room is given by the balance between the generation and removal processes. Jacobi's main concern was uranium mine air, but the same principles and arguments are fully applied to any indoor environment such as private homes and workplaces (Knudson, 1988). The rate of progress of each process is proportional to the concentration of the precursor species for that process. Thus, all the essential information is contained in a set of proportionality rate constants. Assuming complete air mixing and equal ventilation, aerosol-attachment and

deposition rates for all ^{222}Rn and ^{220}Rn decay products, the set of differential equations predicting their indoor concentration dynamics among their possible states are

$$\frac{\partial C_i^{\mathrm{u}}}{\partial t} = \lambda_i C_{i-1}^{\mathrm{u}} + p_{i-1} \lambda_i C_{i-1}^{\mathrm{a}} - \left(\lambda_i + \lambda_{\mathrm{a}} + \lambda_{\mathrm{v}} + \lambda_{\mathrm{d}}^{\mathrm{u}}\right) C_i^{\mathrm{u}} + \lambda_{\mathrm{v}} C_i^{\mathrm{u,out}} \qquad (3.17)$$

$$\frac{\partial C_i^{\mathrm{a}}}{\partial t} = \lambda_{\mathrm{a}} C_i^{\mathrm{u}} + \left(1 - p_{i-1}\right) \lambda_i C_{i-1}^{\mathrm{a}} - \left(\lambda_i + \lambda_{\mathrm{v}} + \lambda_{\mathrm{d}}^{\mathrm{a}}\right) C_i^{\mathrm{a}} + \lambda_{\mathrm{v}} C_i^{\mathrm{a,out}} \qquad (3.18)$$

$$\frac{\partial C_i^{\mathrm{d}}}{\partial t} = \lambda_i C_{i-1}^{\mathrm{d}} + \lambda_{\mathrm{d}}^{\mathrm{u}} C_i^{\mathrm{u}} + \lambda_{\mathrm{d}}^{\mathrm{a}} C_i^{\mathrm{a}} - \lambda_i C_i^{\mathrm{d}} \qquad (3.19)$$

where: the superscripts u, a and d stand for airborne-unattached, aerosol-attached and surface-deposited ^{222}Rn or ^{220}Rn progeny within a reference room, respectively,

C_i^{d} represents the i-th daughter concentration deposited on the available surfaces within the reference room (defined as the ratio of the corresponding activity of the deposited i-th daughter and the considered indoor air volume),

$C_i^{\mathrm{u,out}}$ ($C_i^{\mathrm{a,out}}$) is the outdoor airborne-unattached (aerosol-attached) concentration of the i-th daughter, and

p_{i-1} is the associated recoil factor or the desorption probability of the i-th daughter due to the decay of its attached precursor $i-1$ ($p_{i-1} = 0.8$ in the case of α-emissions and $p_{i-1} = 0$ for β-emissions).

Note that for the noble gases ^{222}Rn and ^{220}Rn ($i = 0$), we have $C_0 = C_0^{\mathrm{u}}$ and $C_0^{\mathrm{a}} = C_0^{\mathrm{d}} = 0$, and that for each ^{222}Rn and ^{220}Rn progeny i, its airborne concentration is then $C_i = C_i^{\mathrm{u}} + C_i^{\mathrm{a}}$.

To well characterise any real situation of indoor air, it must be taken into account that the rate constants of Equations (3.17), (3.18) and (3.19) — excluding the radioactive decay constants, which are true physical constants whose values do not change with circumstances — are highly dependent upon the enclosure geometry, the aerosol concentration, the air movement, the ambient conditions as well as the inhabitant activities and the HVAC systems used. All these effects are time-dependent and vary from one environment to another. For practical reasons, it is not possible to determine experimentally their values from direct measurements in private homes and workplaces. Table 3.1 summarises the typical range of variation and the *baseline* values of indoor ventilation, aerosol-attachment and surface deposition (aerosol-attached and airborne-unattached) rates. The baseline values

are taken from the literature and correspond to the geometric mean of the associated range (ICRP, 1987; Knutson, 1988; Porstendörfer, 1994; UNSCEAR, 2000). According to these data, it is shown that both aerosol-attachment and surface deposition are fast processes for unattached ^{222}Rn and ^{220}Rn progeny. In the case of aerosol-attached ^{222}Rn and ^{220}Rn progeny, the competitive processes are deposition and ventilation, and these are compatible especially at low ventilation rates.

Table 3.1. Typical range of variation and baseline values of the indoor ventilation, attachment and deposition (airborne-unattached and aerosol-attached) rates.

Parameter		Typical values (h^{-1})	
		Range of variation	baseline
Ventilation	λ_v	0.2 - 2	0.7
Aerosol attachment	λ_a	5 - 500	50
Unattached plate-out	λ_d^u	5 - 110	40
Attached plate-out	λ_d^a	0.05 - 1.1	0.4

For practical reasons, the use of baseline values for predicting the average effect over a period of time long enough (some weeks up to a year) is appropriate as a point of departure for calculations. Then, the steady-state solutions of Equations (3.17), (3.18) and (3.19) — neglecting the outdoor airborne-unattached and aerosol-attached concentrations since they are much less than those indoors — that describe the partitioning of ^{222}Rn and ^{220}Rn progeny concentrations within a reference room are given by

$$C_i^u = \frac{\lambda_i(C_{i-1}^u + p_{i-1}C_{i-1}^a)}{\lambda_i + \lambda_a + \lambda_v + \lambda_d^u} \tag{3.20}$$

$$C_i^a = \frac{\lambda_a C_i^u + (1 - p_{i-1})\,\lambda_i C_{i-1}^a}{\lambda_i + \lambda_v + \lambda_d^a} \tag{3.21}$$

$$C_i^d = \frac{\lambda_i C_{i-1}^d + \lambda_d^u C_i^u + \lambda_d^a C_i^a}{\lambda_i} \tag{3.22}$$

To illustrate the effect of changing parameters, Figure 3.2 is presented here as a guide to a general understanding of the portioning of indoor ^{222}Rn and ^{220}Rn decay product concentrations within a reference room. This non-quantitative figure simply attempts to show a subsequent indoor ^{222}Rn or ^{220}Rn progeny behaviour when varying the indoor aerosol concentration. Accordingly, in the absence of aerosol particles the ^{222}Rn and ^{220}Rn daughters may exist in only two states: airborne-unattached and surface-deposited.

As the aerosol concentration is increased the effect of the aerosol-attachment process is to suppress both airborne-unattached and surface-deposited concentration fractions until at very high aerosol concentrations where little concentrations are to be found in either airborne-unattached or surface-deposited states.

It should be noted that the whole concentration of airborne-unattached, aerosol-attached and surface-deposited of the considered daughter never reach the absolute value of the parent — ^{222}Rn or ^{220}Rn — concentration, because certain fraction of them are carried away from the indoor air as a consequence of the ventilation process. The knowledge of ^{222}Rn and ^{220}Rn concentration is adequate in some cases to estimate an upper limit for radiation exposure from their progeny.
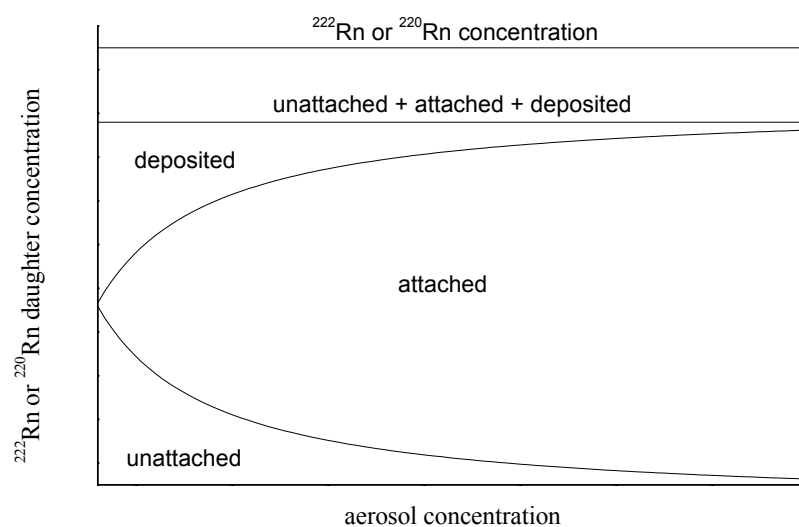

Figure 3.2. Partitioning of indoor ^{222}Rn and ^{220}Rn decay product concentrations as a function of aerosol concentration within a reference room.