

Radon sources and impacts: a review of mining and non-mining issues

Gavin M. Mudd

Published online: 7 September 2008
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Abstract Radon is a ubiquitous natural carcinogen derived from the three primordial radionuclides of the uranium series (^{238}U and ^{235}U) and thorium series (^{232}Th). In general, it is present at very low concentrations in the outdoor or indoor environment, but a number of scenarios can give rise to significant radiological exposures. Historically, these scenarios were not recognised, and took many centuries to understand the links between the complex behaviour of radon and progeny decay and health risks such as lung cancer. However, in concert with the rapid evolution in the related sciences of nuclear physics and radiological health in the first half of the twentieth century, a more comprehensive understanding of the links between radon, its progeny and health impacts such as lung cancer has evolved. It is clear from uranium miner studies that acute occupational exposures lead to significant increases in cancer risk, but chronic or sub-chronic exposures, such as indoor residential settings, while suggestive of health risks, still entails various uncertainties. At present, prominent groups such as the BEIR or UNSCEAR committees argue that the ‘linear no threshold’ (LNT) model is the most appropriate model for radiation exposure management, based on their detailed review and analysis of uranium miner, residential, cellular or

molecular studies. The LNT model implies that any additional or excess exposure to radon and progeny increases overall risks such as lung cancer. A variety of engineering approaches are available to address radon exposure problems. Where high radon scenarios are encountered, such as uranium mining, the most cost effective approach is well-engineered ventilation systems. For residential radon problems, various options can be assessed, including building design and passive or active ventilation systems. This paper presents a very broad but thorough review of radon sources, its behaviour (especially the importance of its radioactive decay progeny), common mining and non-mining scenarios which can give rise to significant radon and progeny exposures, followed by a review of associated health impacts, culminating in typical engineering approaches to reduce exposures and rehabilitate wastes.

Keywords Radon · Radon progeny · Uranium mining · Indoor radon · Health impacts · Radiation exposure

1 Introduction

Radon is a radioactive member of the noble gases, and is derived from the decay of primal uranium or thorium. As an element, radon was first confirmed and studied over the period 1898–1903, and involved

G. M. Mudd (✉)
Department of Civil Engineering, Monash University,
3800 Clayton, Australia
e-mail: Gavin.Mudd@eng.monash.edu.au

many prominent scientists such as Ernest Rutherford, Frederick Soddy, Marie and Pierre Curie, Friedrich Ernst Dorn and others. Despite its somewhat late discovery by science compared to many other elements, radon and its associated health impacts have been felt for many centuries. For example, by the sixteenth century in the Erzgebirge ('Ore Mountains') of central Europe, miners often complained of 'lung-wasting disease' (i.e. lung cancer) but a cause remained elusive and mysterious. It was from mines in this region that uranium was first isolated in 1789 by Martin Klaproth, although health studies suggesting links between uranium, radon exposure and lung cancers would not emerge until a century later.

Throughout the twentieth century, extensive monitoring and research has allowed a more wide-ranging picture of radon and its radioactive decay progeny to emerge, as well as a more comprehensive understanding of potential health issues linked to radon exposure. Radon is considered to be responsible for about half of natural radiation exposure (e.g. UNSCEAR 2000), and therefore possibly a major contributor to background health impacts such as lung cancer (in the absence of other risk factors such as smoking) (e.g. Haque and Kirk 1992; NAS 1999a; Pearce and Boyle 2005; Rosario and Wichmann 2006). The link between sources, exposure and impacts is not always clear or decisive, but the modern approach adopts a generally cautious stance on radon exposures (commonly by minimisation). Since radon is derived directly from the decay of radium, its behaviour can often be governed as much by its parent radium as much by the primary source of uranium (or thorium).

This paper presents a broad review of radon issues. Firstly, it briefly reviews the history of radon, from initial discovery to the more complex understanding of its role in radiological exposures and health impacts. Secondly, it covers the principal physical, chemical and radiological properties of radon, as required for source, transport, exposure or remediation studies. A compilation of natural or background radon is then presented. Next the paper reviews common scenarios for radon sources and exposures, covering non-mining situations such as residential (indoor) radon, caves, and earthquakes, moving to various mining-related issues for uranium, mineral sands, phosphate, oil and gas, gold and coal mining and some miscellaneous problems. This leads to a

discussion of radon exposure and health issues, ending up in different remediation strategies commonly employed to address radon sources and minimise potential exposures. The paper is therefore intended to be a broad but thorough coverage of the principal radon sources, exposures and impacts.

2 Brief history

The effects of radon had been felt by Erzgebirge miners since at least the sixteenth century, as noted by Agricola in his seminal 1556 work *De Re Metallica* (Agricola 1556), though a cause-effect relationship for the sicknesses remained obscure. In 1789, German chemist Martin Klaproth first isolated uranium minerals from these mines (Schneeberg and Joachimsthal) (Habashi and Dufek 2001). Uranium was mined essentially for boutique purposes, such as dyes and ceramic glazes. The discovery and proof of the phenomenon of radioactivity from uranium was made by French physicist Henri Becquerel by mid-1896—starting a rapid revolution in the field soon to become known as nuclear physics (Gowing 1964; Weeks and Leicester 1968; Boorse et al. 1989). Soon afterwards, Becquerel's Polish assistant and research student, Marya Sklodowska (soon to become famous as Marie Curie), and her French husband Pierre Curie isolated the main sources of the radioactivity between 1898 and 1902 as the new elements polonium and radium; also demonstrating in 1898 that thorium was radioactive (Gowing 1964; Habashi 2001).

Around this period, between 1898 and 1903, many scientists were independently researching uranium, thorium and the new phenomenon of radioactivity. In 1899, Ernest Rutherford carefully demonstrated in his laboratory at McGill University in Canada that thorium (ie. ^{232}Th) led to an 'emanation' of radioactive particles—what we now know as 'thoron' or radon-220 (^{220}Rn) (Wilson 1983). Over 1900 to 1902, Friedrich Ernst Dorn in Germany, Frederick Soddy joining Rutherford in Canada and Marie Curie in France all noted that radium emanated radioactive 'particles'—what we now know as radon-222 gas (^{222}Rn). Although Dorn is often given credit as the first to identify the existence of radon, Marshall and Marshall (2003) recently revisited the historical papers and places involved and argued convincingly that due credit for radon's discovery should be given

to Rutherford (and possibly Soddy). The role of water in boosting emanation was also observed. The emanation from actinium ('actinon' or ^{219}Rn) was discovered independently in 1904 by Freidrich Fiesel and André Debierne. Rutherford and Soddy published their seminal papers in 1903 that radioactive decay led to new elements being formed, with the radioactive decay following a simple geometric law leading to a characteristic property or 'half-life' for each radioactive element (Cothorn and Smith 1987). The science of nuclear physics was thus born and continued to evolve rapidly over the coming decades (culminating in the atomic bomb in 1945).

The use of radium emanation as a potential health treatment was proposed soon after its discovery and the medical use of radium, and later radon, quickly accelerated as radium slowly became available (Caufield 1989; Mogren 2002). Throughout the 1910s radium-laced waters were sold as health tonics, radioactive spas were promoted for asserted health benefits and radium even began to be used in luminescent paints for clock dials and even soldiers in the trenches of World War I (Clark 1997). Demand for medical radium escalated dramatically, reaching an incredible price of \$100,000 *per gram* (Habashi and Dufek 2001). Sources of radium were considered extremely rare, and, following in great tradition, a uranium mining boom began to procure prized radium, though the global market was quickly controlled by a handful of mines or even individual countries such as the United States, Belgium and Canada (e.g. Landa 1993; Habashi 2001; Mogren 2002; Mudd 2005). The 1930s saw the emergence of research suggesting links between radon exposure and health impacts such as lung cancers, initially from studies of the Joachimsthal miners but also from strong evidence of health impacts among radium painters (Cothorn and Smith 1987; Jacobi 1993; Clark 1997). This also coincided with increasing understanding of the potential health impacts of excessive radium exposure. In 1934, the International Commission for Radiological Protection (ICRP), proposed the first standard for exposure to radiation—ushering in the era of health physics to minimise and manage impacts. The ICRP standards have evolved over the decades, generally always decreasing as further research comes to light on the relationship between radiation exposure and health impacts.

At the start of World War II, radioactive decay chains were well defined for uranium (^{238}U , ^{235}U) and thorium (^{232}Th), analytical testing was of increasing accuracy and able to detect very low activities, and potential scenarios for exposures and health impacts were beginning to be understood—though much remained to be studied for the latter area of radon.

3 Properties of radon

3.1 Basic properties

Radon is the heaviest member of the noble gas family and is colourless, odourless, relatively chemically inert, naturally radioactive, and has the highest melting point, boiling point, critical temperature and critical pressure of noble gases (Cothorn and Smith 1987). It is soluble in water, with solubility decreasing with increasing temperature, as shown in Fig. 1. An important property of radon is its higher solubility in organic solvents compared to water, a property used in various analytical or field techniques (e.g. Al-Azmi et al. 2004; Fan et al. 2007; Schubert et al. 2007). In general, radon behaves as an inert gas, though it can form clathrates and complex fluorides (no successful formation of oxides or other halides is known) (Cothorn and Smith 1987). The chemistry of

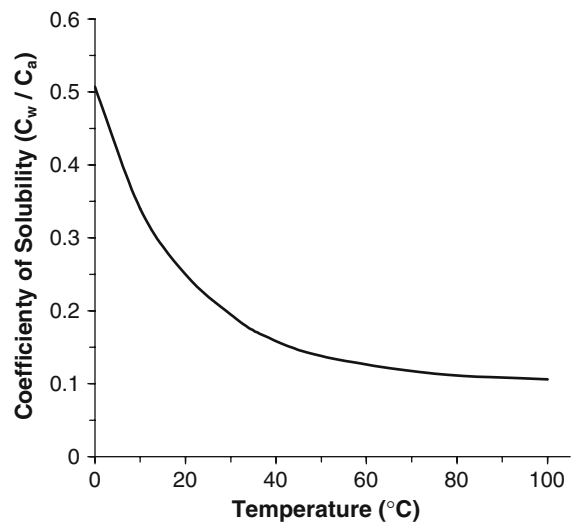


Fig. 1 Solubility of radon in water with respect to temperature (where C_w and C_a are concentrations in water and air, respectively) (adapted from Cothorn and Smith 1987)

Table 1 Uranium (^{238}U), Actinium (^{235}U) and Thorium (^{232}Th) decay chain and weighted average alpha energies (MeV) leading to ^{222}Rn , ^{220}Rn and ^{219}Rn ^a

| | Half-life | α (MeV) | | Half-life | α (MeV) | | Half-Life | α (MeV) |
|-------------------|-------------------|----------------|-------------------|-----------|----------------|--------------------------------|---------------------|----------------|
| ^{238}U | 4.51 Gy | 4.16 | ^{235}U | 710 Gy | 4.20 | ^{232}Th | 14.1 Gy | 4.00 |
| ^{234}Th | 24.1 d | | ^{231}Th | 25.5 d | | ^{228}Ra | 5.77 y | |
| ^{234}Pa | 70.2 s | | ^{231}Pa | 32.5 ky | 4.97 | ^{228}Ac | 6.12 h | |
| ^{234}U | 247 ky | 4.76 | ^{227}Ac | 21.6 y | | ^{228}Th | 1.91 y | 5.40 |
| ^{230}Th | 75 ky | 4.67 | ^{227}Th | 18.5 d | 5.97 | ^{224}Ra | 3.64 d | 5.67 |
| ^{226}Ra | 1600 y | 4.77 | ^{223}Ra | 11.4 d | 5.83 | ^{220}Rn | 54.9 s | 6.29 |
| ^{222}Rn | 3.82 d | 5.49 | ^{219}Rn | 4.01 s | 6.76 | ^{216}Po | 150 ms | 6.78 |
| ^{218}Po | 183 s | 6.00 | ^{215}Po | 1.8 ms | 7.39 | ^{212}Pb | 10.6 h | |
| ^{214}Pb | 0.467 h | | ^{211}Pb | 0.602 h | | ^{212}Bi ^a | 1.01 h | 6.05 |
| ^{214}Bi | 0.328 h | | ^{211}Bi | 129 s | 6.57 | ^{208}Tl | 186 s | |
| ^{214}Po | 164 μs | 7.69 | ^{207}Tl | 286 s | | ^{206}Pb | Stable | |
| ^{210}Pb | 22.3 y | | ^{207}Pb | Stable | | | | |
| ^{210}Bi | 5.01 d | | | | | ^{212}Bi ^a | 1.01 h | |
| ^{210}Po | 138 d | 5.30 | | | | ^{212}Po | 0.304 μs | 8.78 |
| ^{206}Pb | Stable | | | | | ^{208}Pb | Stable | |

^a Approximately 64% of ^{212}Bi decays by α and 36% by β ; other minor decays not included

y, years; d, days; h, hours; s, seconds; G, billion (10^9); k, thousand (10^3); m, thousandths (10^{-3}); μ , millionths (10^{-6})

References: UNSCEAR 1993; Titayeva 1994; IAEA 2003; Appleton 2005; BNL 2008

radon remains relatively understudied compared to other noble gases (Malli 2001).

The element radon has 86 protons and a variable number of neutrons in its atomic nucleus, due to the radon being derived from a different parent radionuclide decay series. The three primary sources for natural radon are the parent isotopes of the two uranium series (^{238}U and ^{235}U) and the thorium series (^{232}Th); with the decay chain sequences and alpha energies shown in Table 1. These chains give rise to the specific radon isotopes of ^{222}Rn , ^{219}Rn and ^{220}Rn , respectively, and their decay products, commonly referred to as ‘progeny’. Although there are 33 radon isotopes known with 110–142 neutrons (Ekström and Firestone 2008), only radon (^{222}Rn), actinon (^{219}Rn) and thoron (^{220}Rn) are relevant in natural or industrial contexts. The extent to which radon, actinon and thoron isotopes (used hereafter to refer to their respective radon isotopes) are present in a given situation will depend on whether the decay chain is in secular equilibrium (i.e. undisturbed) and the primary concentration of uranium and thorium.

All radon isotopes give rise to progeny of polonium, bismuth, tellurium (actinium series only) and

lead with half-lives ranging from fractions of a second (e.g. ^{214}Po) to several years (e.g. ^{210}Pb). These relatively short half-lives give rise to the progeny all having a very high specific radioactivity, and includes numerous alpha, beta and gamma decay steps (see later sections). The behaviour of radon and progeny is therefore critical to understand in order to predict radiation exposures.

The process of alpha decay leads to the recoil of both the alpha particle (which is a charged helium atom, $^4\text{He}^{2+}$) and the progeny. For radon and progeny, this can be crucial in understanding the release into the environment, and will depend on the location of the parent isotope, crystal pore structure and the presence of water, shown in Fig. 2. The recoil distance a radon (^{222}Rn) atom can travel in different media is about 20–70 nm for solids, 100 nm in water and 64,000 nm in air (Tanner 1980; Greeman and Rose 1995).

A final critical aspect of radon and progeny behaviour is the attached–unattached fraction issue. Radon is a relatively inert noble gas but its progeny are all considerably more chemically reactive, leading to an important property for progeny of attaching

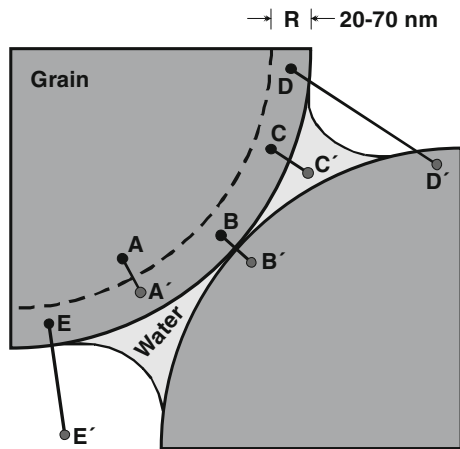


Fig. 2 Radon atom recoils: A–A' inside same mineral grain; B–B' from one mineral to adjacent mineral; C–C' from mineral to water; D–D' from mineral through air to adjacent mineral; E–E' from mineral to air (adapted from Cothorn and Smith 1987; Lawrence 2005)

to aerosols and particulates in the atmosphere. This leads to the fundamental distinction between attached and unattached progeny, a critical aspect in estimating the biological effects of progeny and radiation exposure in lungs (see later health section) (e.g. USDoE 1990; Lugg and Probert 1997; NAS 1999a).

3.2 Units

Due to the complex and rapid evolution in the scientific understanding of radon and progeny, a variety of units have historically been used, especially with respect to assessing exposures and health impacts. For completeness, these include (common symbol, units):

- *Curie*: specific activity of one gram of pure radium-226 (symbol *Ci*);
- *Becquerel*: one radioactive decay per unit time (symbol *Bq*, $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$, $1 \text{ pCi/l} = 37 \text{ Bq/m}^3$);
- *Electron volt*: the product of the charge of an electron and one volt (symbol *eV*; $1 \text{ eV} = 1.6 \times 10^{-19} \text{ J}$);
- *Potential alpha energy*: the total alpha energy emitted by a radon atom as it undergoes complete decay, ideally giving a measure of the energy released if that atom decayed completely inside a lung (symbol *PEA* or ϵ_p , units *MeV per atom*). In practice, since the half-life of ^{210}Pb is 22.3 years

and it would most likely be excreted by lung fluids away from the lung before decay, only the alpha energies from ^{222}Rn to ^{214}Po are included;

- *Potential alpha energy concentration*: the cumulative or total potential alpha energy in a given air volume (symbol *PEAC* or c_p , units *MeV/l* or J/m^3);
- *Equilibrium equivalent concentration*: due to disequilibrium between radon and progeny, this is defined as the activity of the parent radon gas in secular equilibrium which has the same potential alpha energy concentration as the non-equilibrium sample (symbol *EEC*, units Bq/m^3). An equilibrium factor is also defined as the ratio of EEC to the equilibrium PAEC (symbol *F*);
- *Working level*: similar to PAEC, the working level was, initially, somewhat arbitrarily defined as 100 pCi/l (3.7 Bq/l) of air for each of the alpha decays from ^{222}Rn to ^{214}Po , chosen on the belief that this should minimise potential health impacts such as lung cancer (units *WL*, note $1 \text{ WL} = 1.3 \times 10^5 \text{ MeV/l}$). This was subsequently relaxed to any combination of radon and progeny which leads to $1.3 \times 10^5 \text{ MeV}$ of potential alpha energy, and it implicitly assumes a secular equilibrium situation (e.g. $1 \text{ WL} = 3,700 \text{ Bq/m}^3$ ^{222}Rn ; similar activities can be calculated for ^{220}Rn and ^{219}Rn at equilibrium);
- *Potential alpha energy exposure*: given that working level is the activity concentration at a specific time, the cumulative WL over time gives a measure of the total alpha energy exposure (symbol *E*, units *WLM*). Commonly, for convenience in worker health studies, a monthly unit was adopted, giving exposure of ‘working level months’ (assuming say 2000 h of work per year or $\sim 170 \text{ h}$ per month);
- *Absorbed dose*: this is a measure of the radiation energy absorbed directly by cells (symbol *D*, units *J/kg*, named *Gray* or symbol *Gy*);
- *Dose equivalent*: in order to compare effects of radiation, an equivalent unit is required which allows for the differing biological effects and sensitivity of alpha, beta and gamma radiation (symbol *H*, units *Sievert* or *Sv*). The Sievert represents the effective biological impact from radiation exposure after taking into account weighting factors for organ sensitivity, radiation type and other factors.

3.3 Measurement

There are a variety of techniques available to measure radon and progeny. In essence, they can be grouped into three principal techniques: (i) grab sampling; (ii) continuous; (iii) integrative over time (Cothorn and Smith 1987; Harley 1992). Some equipment can be used under either group, such as scintillation cells or ionisation chambers being used for grab sampling or continuous monitoring. Ideally, measurement techniques should establish the respective activities of all radon and progeny isotopes, thereby facilitating the most accurate biological dose models for exposure assessments. However, in practice, the inherent complexity of radon and progeny behaviour means a compromise is required between practicality and theoretical considerations (Cothorn and Smith 1987). The common equipment used for each group includes (e.g. Cothorn and Smith 1987; Harley 1992; IAEA 1992b; Lawrence 2005):

- (i) *Grab sampling*: scintillation cells, ionisation chambers, two filter method;
- (ii) *Continuous*: scintillation cells, ionisation chambers, passive barrier with progeny collection on scintillator, two filter method;
- (iii) *Integrative*: passive barrier with progeny collection on thermoluminescent dosimeter (TLD), activated charcoal, solid state nuclear track detectors (also called “track etch” detectors).

It is important that techniques and programs for radon assessment include calibration and quality control, as there can be wide variability in radon measurements—even using standard equipment and techniques (Djefal et al. 1992). The advent of powerful portable computing and electronics is making more complex field instrumentation practicable (e.g. Todd 1998; Martin et al. 2004; Lawrence 2005), such as linking continuous radon and thoron devices to weather stations. A radon–thoron emanometer for use in measuring field exhalation rates is shown in Fig. 3.

3.4 Exhalation behaviour

The release or escape of radon isotopes and their progeny from its parent radionuclide is a complex process and dependent on many factors. The process, when first recognised by Rutherford, the Curies and



Fig. 3 Radon and thoron emanometer and field laptop (Lawrence 2005)

others, was quickly termed ‘emanation’. For this paper, emanation is used to refer to release from a mineral particle into adjacent pore space, while ‘exhalation’ refers to release into the surface environment.

The location of the uranium or thorium and the respective radium isotopes are fundamental, such as the mineral and its crystal structure (e.g. surface coating of uraninite on a silica grain versus immobilised inside a monazite mineral). Other factors which can influence the exhalation rate include moisture content, barometric pressure, preferential pathways (e.g. cracks, fractures), temperature, particle size and morphology, radium distribution (e.g. diffuse or concentrated, especially with respect to mineral surfaces), seasonal and vegetation effects (e.g. Dyk and Tan 1978; Tanner 1980; Hart 1986; Kvasnicka 1986; IAEA 1992a, b; Schumann and Gundersen 1996; Storm 1998; Storm and Patterson 1999; Lawrence 2005; Schmidt and Regner 2005). It

is unfortunate that many studies on radon fluxes from uranium mining and milling projects, in Australia at least, have not measured or reported most of the above factors, making only cautious comparisons possible (Mudd 2008b).

The fraction of radon which is released relative to its total production is known as the emanation coefficient, and can range from 0 to 1 but is generally between 0.2 and 0.5 (Tanner 1980; Greeman and Rose 1995; Schumann and Gundersen 1996). The emanation coefficient, in turn, is very dependent on moisture content (e.g. Strong and Levins 1982), with an example shown in Fig. 4. In high-grade ores, radiation damage can also increase the emanation coefficient, though this effect is not always present (IAEA 1992b).

Overall, these factors can lead to significant variation in radon exhalation rates and radon and progeny activities in air, such as diurnal variation (e.g. Jackson et al. 1981; Robé et al. 1992; Seftelis et al. 2007), seasonal variation (e.g. Magalhães et al. 2002; Schmidt and Regner 2005; Ruano-Ravina et al. 2008; Zhuo et al. 2008) or possible storm variation due to barometric pressure and/or moisture effects (e.g. Lawrence 2005). It is therefore critically important to understand the principal factors contributing to radon and progeny behaviour at any given site, especially over various time scales.

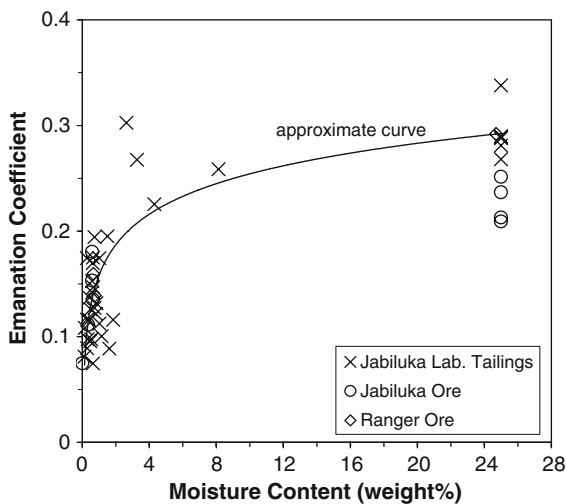


Fig. 4 Effect of moisture content on emanation coefficient for radon from Ranger ore and Jabiluka ore and laboratory tailings (adapted from Strong and Levins 1982; Hart 1986) (25% moisture assumed for saturated samples, based on estimated porosity and density data)

Table 2 Diffusion coefficients and diffusion lengths for radon in various media (Cothorn and Smith 1987)

| Media | Diffusion coefficient (cm ² /s) | Diffusion length (m) |
|------------------|--|----------------------|
| Air | 10 ⁻² | 2.4 |
| Water | 10 ⁻⁵ | |
| Sand | 3 × 10 ⁻² | 1.5 |
| Argillite | 6 × 10 ⁻⁵ | |
| Concrete | 2 × 10 ⁻⁵ | 0.04–0.26 |
| Mineral crystals | 10 ⁻⁹ to 10 ⁻²⁰ | |

As a gas, radon is able to diffuse through different materials, with the diffusive flux proportional to the concentration gradient (Cothorn and Smith 1987). The diffusion coefficient (*D*) will vary according to the media, and can depend on the presence of water, crystal or mineral structure, temperature, radiation damage and particle size distribution (Cothorn and Smith 1987). Typical values for diffusion coefficients in various media are given in Table 2, showing that radon is only likely to migrate reasonable distances in air, water or more porous soils (since inside crystals the time taken for diffusion is longer than the half life).

The steady state exhalation of radon is commonly modelled using Fick’s first law of diffusion in one dimension (e.g. Rogers and Nielson 1981; Rogers et al. 1984; Hart et al. 1986; IAEA 1992b; Ferry et al. 2001, 2002; Dinis and Fiúza 2008). The input data required commonly includes particle size distribution (e.g. sand–silt–clay fractions), dry density, radium activity, soil moisture retention characteristics, diffusion parameters, emanation coefficient (wet and dry) and soil thickness and porosity. The model can then be validated against measured field or laboratory data and used to design and predict the performance of engineered systems to minimise radon fluxes.

The techniques used to measure radon and progeny activity may be able to discern the unattached fraction, depending upon whether they are instantaneous or integrative.

4 Background radon

Uranium and thorium are widespread and exist in very low natural concentrations in soils and rocks,

typically about 3 mg/kg for uranium and 10 mg/kg for thorium (e.g. UNSCEAR 1993; Titayeva 1994; Langmuir 1997). This gives rise to a background radon exhalation from the earth's surface; some thoron exhalation studies are known but remain uncommon. Examples of some country studies include:

- *Australia*: seasonally-adjusted arithmetic mean radon and thoron exhalation from Australian soils is about 22 ± 5 and $1,700 \pm 400$ mBq/m²/s, respectively; the average ²²⁶Ra and ²²⁴Ra soil activities are 28 and 35 Bq/kg, respectively (Schery et al. 1989);
- *China*: area-weighted annual average radon exhalation from Chinese soils is about 30 ± 9.4 mBq/m²/s while ²²⁶Ra soil activity ranged from 17.5 to 115.5 Bq/kg (20 samples only) (Zhuo et al. 2008) (UNSCEAR give Chinese average ²²⁶Ra soil activity as 37 ± 22 Bq/kg; UNSCEAR 1993);
- *France*: radon exhalation from French soils ranges from 10 to 50 mBq/m²/s; the average ²²⁶Ra soil activity is 28 Bq/kg (Robé et al. 1992).

The global radon exhalation from soils ranges from 15 to 23 mBq/m²/s (UNSCEAR 1982). Although there are limited studies on background thoron fluxes, values of approximately 1–2 Bq/m²/s appear typical (UNSCEAR 1993). The radon exhalation gives rise to a general atmospheric background radon which can vary significantly—regionally, seasonally or even diurnally. As such, it is critical to note the weather conditions, time and season of measurements. Some examples of background radon activities in ambient outdoor air are shown in Table 3. In general, an outdoor radon activity of the order of 5–10 Bq/m³ is typical, with an equilibrium factor for progeny ranging from about 0.5 to 0.7 and probably averaging about 0.6 (though more extreme values between 0.2 and 1.0 are known) (UNSCEAR 2000). Background radon activities have also been used to model global atmospheric transport processes, in order to provide a validation of such models independent of climatic inputs (e.g. Zaborowski and Whittlestone 1996).

Radon activities in surface waters and groundwaters are, as one could expect, extremely variable. In general, groundwaters are higher in radon than surface waters (e.g. UNSCEAR 1993; NAS 1999b; Santos et al. 2008), primarily due to slower

Table 3 Ambient outdoor radon activities in air in the United States and Mexico (Bq/m³) (adapted from Gesell 1983; Segovia et al. 2007)

| Region | Period of measurement | Average radon |
|---------------------------------|------------------------|---------------|
| Grants Mineral Belt, New Mexico | November | 22.8 |
| Grand Junction, Colorado | Annual | 27.8 |
| Laguna, New Mexico | June | 18.5 |
| Cincinnati, Ohio | Annual (morning) | 16.3 |
| Cincinnati, Ohio | Annual (afternoon) | 5.1 |
| Argonne, Illinois | Late spring/ summer | 11.1 |
| Socorro, New Mexico | Annual | 8.9 |
| Chester, New Jersey | Annual | 8.1 |
| Lloyd, New York | Summer only | 7.4 |
| Lloyd, New York | March/April | 3.0 |
| Washington, DC | Annual (afternoon) | 4.5 |
| Hawaii | May/June | 1.0 |
| Wales, Arkansas | Annual (afternoon) | 0.7 |
| Kodiak, Arkansas | Annual (afternoon) | 0.4 |
| Mexico | Variable | 13–23 |

movement rates and slightly higher radium from dissolved radium plus radon emanation from aquifer sediments. A major study of 100 public water supply systems in China showed a range from 0.04 to 100 Bq/l radon (Ren et al. 1996). In Mexico groundwaters and water supplies are commonly around 2.2 Bq/l radon but can reach a maximum of 34.2 Bq/l radon (Segovia et al. 2007).

The presence of locally elevated radon activities in surface waters can be used to detect possible groundwater discharge and thereby groundwater–surface water interaction (e.g. Cook et al. 2003; Schubert et al. 2006; Mullinger et al. 2007) or submarine groundwater discharge (e.g. Dulaiova et al. 2007; Lamontagne et al. 2007; Santos et al. 2008).

There is some limited information on the ambient or background radon activities in marine waters. Dissolved radon, commonly around 1 mBq/l, is in deficit relative to radium near the surface (~50 m) due to diffusive losses to the atmosphere, while radon is commonly in excess at the sediment–water interface (Cochran 1992). These results, the radon deficit or excess, can be used to assess oceanic mixing processes. A recent study of the Mediterranean Sea

showed generally low radon activities in its marine waters, ranging between 1 and 20 mBq/l from top to bottom, with wide variation over depths to 3 km (Vaupotič et al. 2008).

As noted for radon exhalation, issues such as seasonal and/or diurnal variation, sampling techniques, and so on need to be carefully considered in understanding background radon processes and activities for any given location or segment of the environment.

5 Non-mining radon issues

In its simplest context, it is possible for radon to accumulate to significant levels wherever there is a radium source and diffusion and transport processes are similar to the half-life for radon. It is principally the longest-lived radon (^{222}Rn) isotope involved, and not actinon or thoron, since these latter isotopes have very short half-lives and it is uncommon for situations to develop that allow build-up and exposure (e.g. UNSCEAR 1993).

To date, significant activities of radon have been observed in cave systems and residential dwellings. Due to their prime significance in population doses, only residential dwellings is reviewed in detail herein. A brief summary of other non-mining radon issues is subsequently presented.

5.1 Indoor radon

Although the potential for biologically significant radon exposures due to mining had been recognised since the early twentieth century, it was not until the 1970s that clear evidence came to light that it was possible for naturally-derived radon (i.e. excluding contaminated sites) to accumulate in residential dwellings, non-uranium mines or other situations to radiologically significant levels (Lugg and Probert 1997; IAEA 2003). Since this time there have been a large number of studies around the world investigating ambient radon activities in residential dwellings, including epidemiological studies for possible related health impacts.

According to an extensive compilation presented by UNSCEAR (2000), given in Table 4, an indoor radon activity of between 30 and 40 Bq/m³ is typical, with an apparent relationship to latitude shown in

Fig. 5. The data suggests that closer to the equator has lower ambient indoor radon, most likely due to greater ventilation associated with higher temperatures, although the scatter at higher latitudes suggests that other factors can also be important.

The accumulation of radon inside residential dwellings is a complex combination of factors and processes, sometimes competing against each other. The major factors involved in determining radon and progeny activities inside a residential dwelling include geology, climate, building materials, design and construction (especially single or multi-storey), building age, barometric pressure effects, and finally lifestyle (e.g. UNSCEAR 1993, 2000; Lugg and Probert 1997; Rosario and Wichmann 2006; Barros-Dios et al. 2007; Denman et al. 2007). Air pressure differences can suck radon into a dwelling or suppress it from entering (e.g. UNSCEAR 2000). Lifestyle aspects often relate to how a dwelling is utilised and can exacerbate or minimise radon issues. For example, Australia is generally considered to have generally low indoor radon due to an open and outdoor lifestyle, compared to cold climate countries where residences are often enclosed for most of the year. Some of these factors are related, such as climate, building design and lifestyle, however, they are not always related in the same manner in different parts of the world.

Some regions naturally contain elevated uranium and/or thorium in soils and rocks, such as granites up to 40 mg/kg uranium, and this can lead to significant radon emanating into and accumulating in dwellings. Based on studies in the UK, Czech Republic, Germany and elsewhere, the most common geological situations giving rise to elevated U/Th are related to granites (Appleton 2007). Alternately, the earthen materials used in construction may contain elevated U/Th, leading to elevated radon (e.g. alum shale in Sweden).

In a handful of mining towns dwellings were sometimes built over uranium mill tailings (or other tailings with elevated radium), or even using tailings in building materials, leading to major radon and progeny exposures for residents in any case (in some cases higher than underground uranium miners). Examples include:

- *Grand Junction and Mesa Counties, Colorado, USA*: approximately 312,000 tonnes of uranium

Table 4 Global summary of indoor radon (^{222}Rn) activities (Bq/m^3) (UNSCEAR 2000), including additional Mexican data from Segovia et al. (2007) and Paarl, South African data from Lindsay et al. (2008)

| Region | Country | Arithmetic mean | Geometric mean | Maximum | Geometric SD |
|-----------------|----------------------|-----------------|----------------|---------|--------------|
| Africa | Algeria | 30 | | 140 | |
| | Egypt | 9 | | 24 | |
| | Ghana | | | 340 | |
| | South Africa (Paarl) | ~37–132 | | 465 | |
| North America | Canada | 34 | 14 | 1720 | 3.6 |
| | United States | 46 | 25 | | 3.1 |
| Central America | Mexico | 83 | 75 | 448 | |
| South America | Argentina | 37 | 26 | 211 | 2.2 |
| | Chile | 25 | | 86 | |
| | Paraguay | 28 | | 51 | |
| East Asia | China | 24 | 20 | 380 | 2.2 |
| | Hong Kong | 41 | | 140 | |
| | India | 57 | 42 | 210 | 2.2 |
| | Indonesia | 12 | | 120 | |
| | Japan | 16 | 13 | 310 | 1.8 |
| | Kazakstan | 10 | | 6000 | |
| | Malaysia | 14 | | 20 | |
| | Pakistan | 30 | | 83 | |
| | Thailand | 23 | 16 | 480 | 1.2 |
| West Asia | Armenia | 104 | | 216 | 1.3 |
| | Iran | 82 | | 3070 | |
| | Kuwait | 14 | 6 | 120 | |
| | Syria | 44 | | 520 | |
| North Europe | Denmark | 53 | 29 | 600 | 2.2 |
| | Estonia | 120 | 92 | 1390 | |
| | Finland | 120 | 84 | 20000 | 2.1 |
| | Lithuania | 55 | 22 | 1860 | |
| | Norway | 73 | 40 | 50000 | |
| | Sweden | 108 | 56 | 85000 | |
| West Europe | Austria | | 15 | 190 | |
| | Belgium | 48 | 38 | 12000 | 2.0 |
| | France | 62 | 41 | 4690 | 2.7 |
| | Germany | 50 | 40 | >10000 | 1.9 |
| | Ireland | | 37 | 1700 | |
| | Luxembourg | 110 | 70 | 2500 | 2.0 |
| | Netherlands | 23 | 18 | 380 | 1.6 |
| | Switzerland | 70 | 50 | 10000 | |
| | United Kingdom | 20 | | 10000 | |
| Eastern Europe | Bulgaria | | 22 | 250 | |
| | Czech Republic | 140 | | 20000 | |
| | Hungary | 107 | 82 | 1990 | 2.7 |
| | Poland | 41 | 32 | 432 | 2.0 |
| | Romania | 45 | | 1025 | |
| | Slovakia | 87 | | 3750 | |

Table 4 continued

| Region | Country | Arithmetic mean | Geometric mean | Maximum | Geometric SD |
|-----------------------------|-------------|-----------------|----------------|---------|--------------|
| South Europe | Albania | 120 | 105 | 270 | 2.0 |
| | Croatia | 35 | 32 | 92 | |
| | Cyprus | 7 | 7 | 78 | 2.6 |
| | Greece | 73 | 52 | 490 | |
| | Italy | 75 | 57 | 1040 | 2.0 |
| | Portugal | 62 | 45 | 2700 | 2.2 |
| | Slovenia | 87 | 60 | 1330 | 2.2 |
| | Spain | 86 | 42 | 15400 | 3.7 |
| Oceania | Australia | 11 | 8 | 420 | 2.1 |
| | New Zealand | 20 | 18 | 90 | |
| Median | | 46 | 37 | 480 | |
| Population-weighted average | | 39 | 30 | 1200 | |

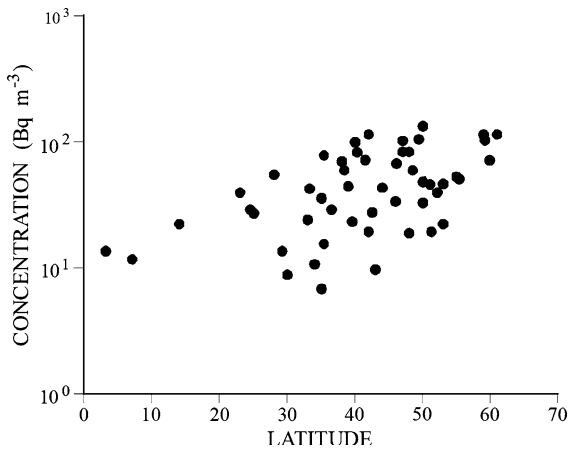


Fig. 5 Average indoor radon activity versus latitude (UN-SCEAR 2000)

mill tailings were used in construction materials throughout the vicinity for more than 4,000 houses, schools, churches, public and commercial buildings (the material was provided freely by the mill) (Hazle et al. 1982; Rael 1999);

- *Canonsburg, Pennsylvania, USA*: the site of both a radium refinery in the early twentieth century and later a uranium mill from 1942 to 1957, Canonsburg also saw some mill tailings taken from the site for construction purposes (USDoe 2001);
- *Eastern Germany (former GDR)*: waste rock from the former Crossen uranium mine was used in buildings in eastern Germany (Küppers and

Schmidt 1994); another survey of 1700 homes in eastern Germany returned activities up to 15,000 Bq/m³ with one extreme value of 115,000 Bq/m³, with more than 50% of homes at Schneeberg greater than the local action limit of 250 Bq/m³; (Vandenhove et al. 2006);

- *South Africa*: similarly to the USA and Germany, local communities in the south-west Karoo Province used stockpiled uranium ore for road construction or farmhouse foundations, leading to indoor radon activities of 351–835 Bq/m³ and exposures of 6.0–14.2 mSv/year (Scholtz et al. 2005);
- *Hunters Hill, inner suburban Sydney, Australia*: the site of an old radium extraction refinery (1911–1915) and adjacent tin smelter (1895–1964) was redeveloped for residential housing though the site has yet to be satisfactorily remediated (Mudd 2005).

In areas around the world known for elevated indoor radon, there are often specific building codes, regulations or guidelines to ensure that building designs and construction minimise radon build-up and associated radiation exposures (e.g. USEPA’s “Citizen’s Guide”, which suggests an action level of 148 Bq/m³ for indoor radon; USEPA 2007).

Indoor radon and progeny activities are rarely in equilibrium, typically showing an equilibrium factor of about 40% (NAS 1999a). The differences between the use of short- and long-term detectors for the study of indoor radon been shown to be minimal, though it

is considered more thorough to use long-term detectors (Ruano-Ravina et al. 2008).

A case study of indoor equilibrium-equivalent radon and thoron progeny activities in 10 rammed earth and 10 conventional dwellings at Margaret River, just south of Perth in Western Australia, was given by Walsh and Jennings (2002). The study showed that mean indoor radon and progeny activities were 24 and 9.3 Bq/m³ EEC, respectively, while thoron and progeny activities were 3.9 and 0.8 Bq/m³ EEC, respectively, leading to a combined radiation dose for each dwelling type of 4.1 and 2.2 mSv/year, respectively—significantly above average Australian background radon exposures of about 0.7 mSv/year (see Webb et al. 1999). A more extreme example from Slovenia involved a dwelling with a radon exposure of 9–35 mSv/year (the cause of the high radon is not stated) (Zmazek and Vaupotič 2007).

An online radon and progeny dose calculator has been implemented by Diehl (2008a), based on unit conversion factors (e.g. alpha energy to WL to WLM). Based on a typical indoor radon activity of 40 Bq/m³, equilibrium factor of 0.4 and 60% occupancy over 70 years, it is possible to estimate a natural indoor radon exposure of about 0.53 mSv/year or 0.13 WLM/year.

5.2 Miscellaneous radon issues

A number of other scenarios are known to involve elevated radon activities, radiation exposures or uses. These include:

- *Mineral spas and thermal waters*: such waters often contain appreciable radon (by choice or otherwise). Examples include the Polichnitos hot springs in Greece with radon commonly between 110 and 220 Bq/l (Vogiannis et al. 2004), thermal springs of northern Venezuela with 1–578 Bq/l radon (Horváth et al. 2000), and Paralana spring in South Australia at 1,800–5,800 Bq/l radon (ambient air is highly variable but can range from 1,705 to 10,952 Bq/m³ directly over the two spring pools; radium is 14–17 Bq/l in pool waters) (HR 1998; Brugger et al. 2005).
- *Earthquakes or other land movements*: it has long been recognised that elevated radon activities in soils occur just before an earthquake (e.g. Singh

et al. 1999; Yang et al. 2005; Amponsah et al. 2008), generally considered to be related to the rapid migration of gases just before the earthquake. In some cases, however, seismic activity appears to give rise to reduced radon activities in soils, as measured in Japan (Yasuoka et al. 2005) and Taiwan (Kuo et al. 2006). Recently, radon in soil and/or groundwater has been shown to be linked to tectonic controls adjacent to landslide activity in the Himalayas of northern India (Ramola et al. 2007), as well as subsidence due to former underground iron ore mining in the Luxembourg Basin in Europe (Kies et al. 2006). Field studies at Yerevan, Armenia, have shown that the increases in radon activities due to seismic activity are important in public radiation exposure doses (Saghatelyan et al. 2005).

- *Caves*: underground caves, most commonly in limestone, can also give rise to scenarios of elevated radon and progeny (e.g. (Szerbin 1996; Madden 1997). For example, numerous caves were assessed for radon activities in the mid-1990s in Australia by Solomon et al. (1996). Monitoring data in winter and spring showed radon activities of 500 ± 40 and 795 ± 50 Bq/m³, respectively, with a maximum of 6,330 Bq/m³. In winter 19% of measurements were above the action level of 1000 Bq/m³, rising to 29% for spring. Seasonal variability in radon activities does appear to be common in caves, including equilibrium factors.

Further papers on non-mining radon issues can be found in several recent conferences on naturally occurring radioactive materials ('NORM') held by the International Atomic Energy Agency in Vienna in September 1999 (IAEA 2002), May 2004 (IAEA 2005) and December 2004 (IAEA 2006), amongst many other conferences and considerable literature.

6 Mining radon issues

Mining is the most common industry where the potential for acute exposures can occur, principally in uranium mining but also for other commodities. This section will review several sectors of the mining industry which are known to encounter significant radon and related exposure issues.

6.1 Uranium mining and milling

The mining and milling of uranium ore can lead to high radon exposures, sometimes extremely high if minimal or no precautions are adopted. A typical uranium mine is similar to any other mine, consisting of an underground or open cut mine, economic ore, low grade ore and/or waste rock stockpiles, a processing mill (such as grinding, leaching, solvent extraction, chemical precipitation and calcining) and finally a tailings dam. Other infrastructure may also be required, such as water management facilities, a power station, dedicated road and air transport infrastructure and possibly a nearby town to accommodate workers and their families. A minor amount of uranium is also produced by non-conventional in-situ leach or ‘solution mining’, whereby acidic or alkaline chemical solutions are injected into and extracted from the porous uranium orebody using boreholes (i.e. essentially a chemical solution mining exercise in groundwater; see Mudd 2001a, b).

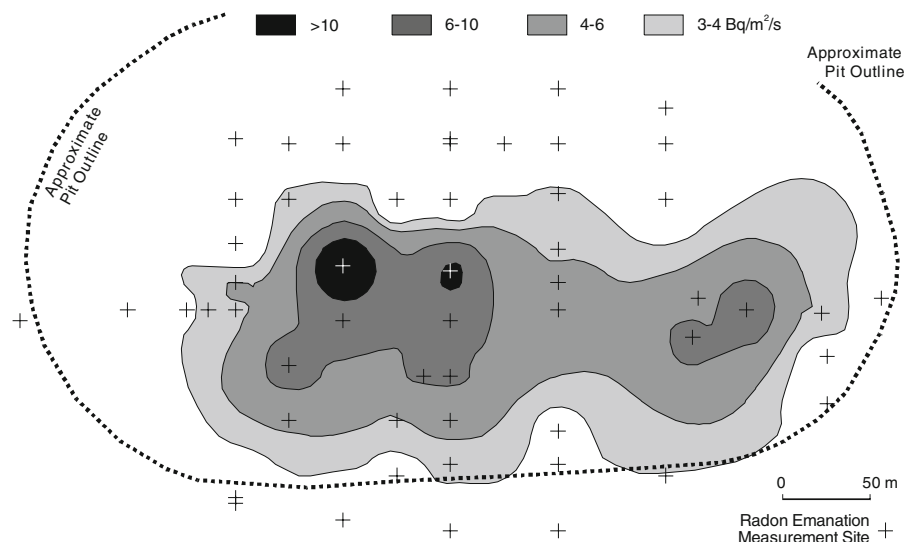
It is possible to seek elevated radon fluxes during uranium exploration programs (e.g. Stewart 1968; Smith et al. 1976; Severne 1978), however, whether there is a notable expression of radon from a uranium deposit at the surface is highly variable. A recent analysis of Australia showed that uranium ore-related radon fluxes were detectable for the Ranger, Yeelirrie, Nabarlek, Koongarra and Lake Way uranium deposits as these all outcrop or subcrop (sometimes over large areas as at Ranger and Yeelirrie), while

other buried uranium deposits such as Olympic Dam, Beverley, Honeymoon and Jabiluka showed no clear deposit-related radon signature (Mudd 2008b). The baseline radon flux contours for the Koongarra 1 uranium deposit are shown in Fig. 6 (Koongarra remains undeveloped).

Mining is commonly the occupation with the highest potential for radon exposures, principally in underground mining, while mill workers and other roles encounter lower radon activities and exposures. The nature of the exposure is complex, and depends on uranium ore grade, deposit mineralogy and geochemistry, ventilation regime (especially open cut versus underground), temperature and the extent and nature of particulates in the mine atmosphere (since this critically affects the attached–unattached progeny fraction and lung dosimetry; Wasiolek and James 2000). Additionally, the older the mine the higher its radon emissions are likely to be (Jackson et al. 1981). Blasting appears to temporarily increase radon and progeny activities as well as alter the equilibrium factor (Ertle et al. 1981; Warneke and Sontner 1989).

The uranium mines of the first half of the twentieth century, operated primarily for radium extraction, included a mix of open cut and underground mining, though it appears that underground mining was more dominant (similar to gold and other mining of the day). Efforts to minimise radiation exposures due to radon and progeny were minimal, as there was still only limited understanding of the links between high

Fig. 6 Pre-mining radon exhalation measured at the Koongarra 1 uranium deposit, 1978 (redrawn from Davy et al. 1978)



exposures and health impacts such as lung cancers (Jacobi 1993).

Following the advent of the Cold War nuclear weapons race from 1945, uranium mining became a major boom industry across the world, especially in the United States, Canada, South Africa, eastern Germany (the former German Democratic Republic, GDR) and many states of the Soviet Union. The first two decades were primarily concerned about urgent uranium production for the nuclear weapons programs of the day—although there was arguably limited but evolving understanding of radiological exposure issues, this came second to uranium production (Jacobi 1993). By the late 1960s, however, radiological exposure issues and standards had become more critical, with increasing attention being given to ventilation to address dust, radon and progeny issues with respect to health impacts (see next section).

There is an extensive range of technical reports, journal papers and conference proceedings with data on radon and progeny activities for uranium mining since the late 1940s. The major groups include the International Atomic Energy Agency (IAEA), International Commission on Radiological Protection (ICRP), Biological Effects of Ionising Radiation (BEIR), United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), numerous government research agencies and regulators as well as technical societies, amongst others.

To illustrate the changes in radon exposures over time, a case study for Australian uranium mining and milling was developed and is shown in Table 5. This case study could be expected to be similar or representative of many other uranium mining countries.

Another major source of radon emissions or exposures from a uranium mining-milling project is tailings, the finely ground rock remaining after chemical processing and uranium extraction. Uranium mill tailings typically retain most of the radium from the original ore (and other radionuclides, including residual uranium), and thus constitute a major radon source term (Clements et al. 1978; IAEA 1992a). Tailings are invariably pumped as a slurry to an engineered storage dam, with the facility decommissioned and rehabilitated upon mine closure. The management of tailings is therefore critical with respect to radon (IAEA 1992b). A brief compilation

of radon fluxes from uranium mill tailings is shown in Table 6.

One approach which has been favoured by some in the uranium industry is the use of water covers, though their effectiveness remains the subject of some conjecture (Mudd 2008b). In Australia water covers have been viewed favourably, especially in the tropics for the operating Ranger and closed Nabarlek uranium projects, however, field evidence of the effectiveness of water covers in reducing radon fluxes and loads is lacking, with theoretical estimates varying widely (Mudd 2008b). Field studies in Brazil have shown that approximately one third of the radon in mine water retention ponds is released to the atmosphere (Paschoa and Nóbrega 1981). Based on laboratory column studies, Rogers and Nielson (1981) argued that the water covers on mill tailings facilities were a major radon source, and presented a model to estimate such releases (implemented online by Diehl 2008c).

The rehabilitation of uranium mill tailings generally involves dewatering (to the best extent practicable) followed by construction of an engineered soil cover over the tailings. The soil cover is designed with multiple layers to ensure some moisture retention and thereby retard radon diffusion, leading to lower radon exhalation at the surface (e.g. Rogers et al. 1984; IAEA 1992b). In Australia, the major public inquiry into the Ranger uranium project (1975–1977) (Fox et al. 1977) recommended final in-pit tailings disposal and management, primarily due to concerns over long-term radon exhalation close to indigenous communities after rehabilitation (Haylen 1981). If the tailings were deep in the former pit and below the water table, then the radon exhalation at the surface would be minimised after final rehabilitation.

A comprehensive review of radon exhalation and loads from uranium mill tailings, economic ore, low-grade ore and waste rock stockpiles and processing mills for numerous Australian uranium projects is presented by Mudd (2008b). The normalised radon released per tonne of uranium oxide production (ie. GBq/t U_3O_8) is variable, with estimates for Australia commonly ranging from 37 to 155 GBq/t U_3O_8 , with one extreme estimate at 2,162 GBq/t U_3O_8 (Mudd 2008b). The US Nuclear Regulatory Commission estimated the normalised radon release for a ‘generic’ uranium mill of about 318 GBq/t U_3O_8 (USNRC 1980). Based on written advice from various uranium

Table 5 Summary of radon activities over time for some Australian uranium mines¹ (compiled from Stewart 1963^a, Rosen 1987; Sonter 1987; Sonter and Hondros 1988; Warneke and Sonter 1989; Woodward et al. 1991; Kinhill 1997; HR 2003–2007; Mudd 2008a)

| 1950–1960s (general) | Typical ore Grade (%U ₃ O ₈) | Radon Average | (Bq/m ³) Maximum |
|---|---|---|---|
| Open cut mines | 0.17–0.35% | 40 | 190 |
| Underground mines—low grade | ~0.1% | 3,000 | 9,300 |
| Underground mines—high grade ^b | 0.3–2.5% | ‘0’–85,000 | 1,110,000 ^c |
| Chemical treatment plants | ~0.7% | – | <190 |
| Code of Practice (1955) | | | 3,700 |
| | Grade (%U ₃ O ₈) | Radon (Bq/m ³) | Progeny (m WL) |
| <i>Radium Hill</i> (underground mine) | | | |
| Before March 1955 | ~0.1% | 2,100–18,000 | 600–1,800 |
| March 1955–1961 | ~0.1% | 100–7,900 | 100–550 |
| <i>Ranger</i> (Nov. 1984–Oct. 1985) (open cut mine) | | | |
| Controlled and supervised areas | ~0.3% | 3,223 samples <3,100 | Maximum 86; 1,020 samples <17 |
| Non-controlled and non-supervised areas | ~0.3% | 1,709 samples <3,700; maximum 1,295; 1,230 samples <185 | Maximum 17.6; mean mine office 4.9; other areas 1.4–4.5 |
| Limits—Controlled / supervised areas | – | 100,000 | 330 |
| Limits—Non-controlled/Non-supervised areas | – | – | 10 |
| <i>Olympic Dam</i> (mid-1980s to late 1990s) ^d | | | |
| Underground ore stockpiles (in drive) | ~0.1% | ~200 | ~110–160 |
| Underground drives and tunnels | ~0.1% | – | ~50–140 |
| Underground open stopes (mid-1980s) | ~0.1% | ~5,200 | ~10,000–20,000 |
| Underground stopes (late 1980s) | ~0.1% | ~2,000 to ~5,000 | ~100–300 |
| Underground stopes—after blasting (late 1980s) | ~0.1% | ~2,000 to ~18,000 | ~200–1,200 |
| Underground radon progeny exposures (1983–86) | Range: 0.2–1.0 WLM; Average ~0.35 WLM | | |
| Underground mine (‘Purple Stope’, mid-1990s) | ~0.08% | – | 5.3–73 |
| <i>Beverley</i> (2000s) | | | |
| ‘Environmental’ radon ^e | – | ~20 to ~195 | ~1 to ~7 |

¹ Uranium projects of the 1950–1960s included Rum Jungle (open cut), Upper South Alligator Valley (open cut, underground), Mary Kathleen (open cut), Radium Hill (underground), Port Pirie (metallurgical plant)

^a Stewart (1963) used units for radon of both ‘ $\mu\text{Ci/l}$ ’ and pCi/l. Based on the paper and its timing, it appears that units of pCi/l are intended and have been assumed above

^b A small stope in a high grade underground mine (unnamed) averaged 5,550 Bq/m³ (ranging up to 22,200 Bq/m³), and progeny concentrations were 0.43×10^5 MeV/l under natural ventilation and 0.125×10^5 MeV/l under forced ventilation of 28.3 m³/min (Stewart 1963)

^c Described as an ‘exceptional value’ by Stewart (1963)

^d These values are estimates and not actual monitoring data (see Sonter 1987)

^e Location of monitoring point in relation to the in-situ leach processing plant unstated, though for some years it is stated as the nearby accommodation camp (see HR 2003–2007)

Note: No data on ambient radon activities appears to be published within active mining areas for the Nabarlek and Mary Kathleen uranium projects

Table 6 Brief survey^a of radon fluxes from uranium mill tailings around the world (IAEA 1992b)

| Mine | Description | Ore grade (%U ₃ O ₈) | Radium (Bq/kg) | Radon Flux (Bq/m ² /s) |
|--------------------------------------|-----------------------------|---|--------------------|-----------------------------------|
| Olympic Dam, Australia | Moisture ~19% | 0.08% | 8,000 | 1.3 |
| Lacnor, Elliot Lake, Canada (closed) | Frozen surface | ~0.12% | 600 to 12,700 | 0.03–1.52 |
| | Vegetated surface | | | 0.31–4.96 |
| | Dry surface | | | 0.20–0.84 |
| | Damp surface, no vegetation | | | 0.18–6.67 |
| | Saturated surface | | | 0.01–0.11 |
| Key Lake, Canada | Very wet | ~2.95% | 100,000 to 315,000 | 0.78, 0.88 |
| | Dry | | | 1.11–10.2 |
| | Damp | | Average to 300,000 | 2.4, 8.6 |
| | Frozen | | | 0.14 |
| Hamr, Czech Rep. | Very wet | 0.12% | – | 0.22 |
| Rössing, Namibia | <1% moisture | 0.035% | 1,000 to 4,000 | 0.9 |
| | 1–10% moisture | | | 0.65 |
| | >10% moisture | | | 0.4 |
| Andujar, Spain | Uncovered | 0.12% | 12,800 | 10 |
| | Clay cover | | | 0.08 |
| | Soil, sand, clay cover | | | 0.024 |
| Western areas, USA | Dry, no cover | 0.1–0.3% | 15,000 to 30,000 | 10–40 |
| | Dry, clay–silt cover | | | 0.3–0.7 |

^a A detailed compilation for Australian uranium mill tailings is given by (Mudd 2008b)

mines, UNSCEAR estimated normalised radon releases for numerous uranium mines in Canada, Australia and eastern Germany (GDR) ranging from 1.2 to 1800 GBq/t U₃O₈, averaging approximately 13 GBq/t U₃O₈ (UNSCEAR 1993). There does not appear to be any major difference in normalised radon releases between underground, open cut or in-situ leach projects, although the UNSCEAR estimates and others available appear to be very crude and approximate.

Uranium mining gives rise to major sources of radon loads and activities, which in turn can give rise to major exposure scenarios for workers or nearby communities. Modern uranium mine design and operation requires substantive effort to address radon issues, and requires constant monitoring and vigilance to ensure compliance with radiation exposure standards.

6.2 Mineral sands mining and milling

Mineral sands mining processes beach or placer sands for the recovery of various heavy minerals such as rutile (TiO₂), ilmenite (FeTiO₃), zircon (Zr(SiO₄))

and monazite (a rare earths-phosphate mineral, e.g. (Ce, La)PO₄) (Lottermoser 2007). The proportion of each mineral in the heavy mineral fraction is variable across deposits. Although monazite typically comprises 1% of the heavy mineral fraction in Australian mineral sands resources, monazite itself contains radiologically significant impurities of thorium (²³²Th, up to several percent) and uranium (Mason et al. 1988). Monazite from Guarapari, Brazil, contains 8–12% ThO₂ (Cockell et al. 2007).

The separation processes applied to heavy mineral sands leads to concentration of the radioactive monazite into a specific concentrate, potentially giving rise to significant radiation exposure issues due to thoron, dust, gamma radiation (due to ²¹⁴Bi) as well as radon. A compilation of the thorium and uranium content of Australian and Brazilian mineral sands fractions is given in Table 7.

In Western Australia in the early 1990s, about 30 million tonnes of material was mined annually, typically containing up to 60 mg/kg thorium and 20 mg/kg uranium (Hewson and Upton 1996). After processing, individual waste streams could contain thorium ranging from 100 to 30,000 mg/kg

Table 7 Concentration ranges of thorium (^{232}Th) and uranium (^{238}U) in Australian and Brazilian heavy mineral sands fractions (Bq/kg) (UNSCEAR 1993; Malanca et al. 1998)

| | Australia ^{232}Th | Australia ^{238}U | Brazil ^{232}Th | Brazil ^{238}U |
|---------------------------|--------------------------------|-------------------------------|-----------------------------|----------------------------|
| Ore | 60–200 | 40 | – | – |
| Heavy mineral concentrate | 1,000–1,300 | <100 | 2,900–60,000 | 480–4,000 |
| Ilmenite | 600–6,000 | <100–400 | 1,765 | 461 |
| Leucoxene | 1,000–9,000 | 250–600 | – | – |
| Rutile | <600–4,000 | <100–250 | – | – |
| Zircon | 2,000–3,000 | 200 to 400 | 473 | 4,409 |
| Monazite | 600,000–900,000 | 10,000–40,000 | 187,00–196,800 | 12,090–13,760 |
| Xenotime | 180,000 | 50,000 | – | – |
| Average soil or rock | 40 | 40 | – | – |

(i.e. 0.01–3% thorium), though the more highly radioactive waste streams were generally smaller in mass. At Byron Bay, on the central coast of New South Wales, Australia, the tailings from mineral sands mining and processing, including low grade monazite concentrates, were used in various low-lying areas of the township for urban re-development, leading to above background radiation exposures in some residences (Gandy and Colgan 1983). A similar case study is known for Capel in Western Australia (King et al. 1983).

6.3 Phosphate mining and milling

Phosphate ore often contains elevated levels of uranium compared to typical soils (thorium is similar to soils), though it is variable from 30 Bq/kg for Kovdor, Russia, to 1,500 Bq/kg in Florida, USA (equivalent to 2.4–121 mg/kg, respectively) (UNSCEAR 2000). The extraction of uranium as a by-product from phosphate mining has been minor but perhaps important, probably of the order of less than 20% of cumulative global uranium production (Mudd and Diesendorf 2008).

Radon issues primarily arise with the processing wastes, mainly phosphogypsum, as well as any use of by-products or other wastes (including liquid wastes). The ^{238}U decay chain is usually close to secular equilibrium, with the radium (^{226}Ra) activity of phosphogypsum typically about 900 Bq/kg (depending on the origin of the ore) (UNSCEAR 1993). The management of associated radon issues in phosphate mining and processing will therefore depend on various site specific factors, such as ore grade,

climate, waste management practices, solid waste use, and so on (e.g. phosphogypsum use in building materials). Estimates of the radon released by typical phosphate projects are 820 GBq/year for a 0.7 Mt/year phosphoric acid facility and 221 GBq/year for a 375 kt/year fertilizer facility (UNSCEAR 2000).

6.4 Oil and gas extraction and processing

The extraction and processing of oil and gas represents a potentially major global radon source term, due mainly to the volumes produced and consumed. The activity of radium, and thereby radon and progeny, is highly variable in oil–gas projects across the world and can be expected to be closely linked to the source petroleum field, though published data is not widespread. During processing, the major risks are the exhalation of radon and buildup of radium in pipe scale. Radium scale is particularly important due to the decay chain leading to bismuth (^{214}Bi), which is a strong gamma emitter. Estimates of the radon released by typical oil–gas facilities are 540 GBq/year for a 3.5 Mt/year oil facility, 500 GBq/year for a 72 Gm³/year gas facility, while a 400 MWe gas-fired power plant is estimated to release 230 GBq/year of radon (UNSCEAR 2000).

Reed et al. (1991) presented data on radium buildup in North Sea and Louisiana oil–gas production facilities:

- *Pipe scales*: 26,000–286,000 Bq/kg;
- *Oil production separator sludges*: 1,000–823,000 Bq/kg;
- *Gas separator sludges*: 2,000–19,000 Bq/kg;
- *Gas separators*: 200–55,000 Bq/kg.

In Poland, oil–gas brines contain a maximum of 258 Bq/l radium (^{226}Ra) (Skowronek et al. 2005). In offshore petroleum exploration near Darwin, Australia, pipe scale contained ^{226}Ra – ^{228}Ra activities up to 600,000 Bq/kg (Cassels and Waite 2001). A study of oil–gas facilities in Nigeria has shown elevated gamma radiation ranging from 0.1 to 15 $\mu\text{Sv/h}$, with radium activity measured in scale up to 200,000 Bq/kg (Elegba and Funtua 2005).

6.5 Gold mining and milling

Commonly, there is only background concentrations of uranium or thorium in most gold ores, with consequent radiation exposures being low and minor in comparison to other occupational health issues in gold mining (e.g. silicosis). There are some exceptions, however, most notably being the gold–uranium ores of South Africa.

Since the discovery and development of the gold fields of South Africa in the late nineteenth century, more than six billion tonnes of gold ore has been mined and processed to produce $\sim 51,000$ tonnes of gold (Mudd 2007), including about 765 million tonnes of ore grading $\sim 0.022\%$ U_3O_8 which was processed to produce about 175,000 tonnes U_3O_8 (Mudd and Diesendorf 2008). The low-grade uranium content of most of the gold–uranium tailings, left almost entirely as tailings on the surface and adjacent to major populations, has led to a major and ongoing radiological exposure issue (Lindsay et al. 2004; Tsela and Zituta 2006). Additionally, there have been major issues with regards to radiation exposure for gold mine workers, including underground miners and those in the mills.

A recent study of Ghana found mean radon activities in underground gold mines of 350–445 Bq/m^3 , leading to exposures of about 1.83 mSv/year (mining only) (Darko et al. 2005). Surface outdoor radon activities ranged from 24 to 41 Bq/m^3 in the mill, tailings dams, stockpiles and shafts/declines, leading to exposures of about 0.13–0.17 mSv/year . Uranium (^{238}U) and thorium (^{232}Th) concentrations were low and essentially background.

6.6 Coal mining and combustion

As with oil and gas, the presence of elevated uranium (or thorium) is highly variable across coal fields

around the world. There are a very small number of coals in the world which contain uranium at potentially economic grades, such as the former Freital coal mine in eastern Germany at 0.1–0.3% U_3O_8 , the dormant Okrzeszyn coal mine in Poland at 0.04–0.12% U_3O_8 , or the dormant Belskoie coal mine in Russia at 0.04–0.12% U_3O_8 (IAEA 1996). Although uranium-rich coal deposits have not been widely processed in the past for their uranium content, when combusted in coal-fired power stations they can cause major releases of radionuclides to the surrounding region as well as enriching the residual fly ash in radionuclides.

In Poland, hard rock coals contain a maximum of 159 Bq/kg radium (^{226}Ra), while associated waste rock contains 122 Bq/kg radium (Skowronek et al. 2005). Pond sediments and pipe scales at coal mines contained radium up to 157,000 Bq/kg radium. A survey of underground coal mines in the United Kingdom gave radon activities of 27–1,244 Bq/m^3 , with radon progeny being 4–40 mWL , showing significant variability between and within mines (Page and Smith 1992). In New South Wales, Australia, coal ash has ^{226}Ra ranging from 88 to 370 Bq/kg, ^{238}U from 70 to 167 Bq/kg and ^{228}Th from 91 to 261 Bq/kg; ^{222}Rn very low at 2.1–6 $\text{mBq/m}^2/\text{s}$ while ^{220}Rn was 20–86 $\text{mBq/m}^2/\text{s}$ (Zahorowski et al. 1994)—all considerably lower than average Australian background fluxes. At the Figueira coal mine in southern Brazil, within a geologic province containing known uranium deposits, radon activities averaged about 1,700 Bq/m^3 (range 200–6,100 Bq/m^3), leading to exposures estimated at 2.1 WLM (range 0.2–7.2 WLM) (Veiga et al. 2006). As such, any assessment of radon and progeny issues associated with coal mining needs to be undertaken on a site-specific basis.

6.7 Other mining

A recent survey of a large, long abandoned tin mine in Cornwall, UK, from which a very small amount of uranium ore was also extracted a century ago (for radium), showed radon activities reaching as high as 3,932,920 Bq/m^3 some 52 m from the tunnel entrance (claimed as one of the highest radon measurements ever recorded in Europe)—with radon activity at 1 m height still being 2,154,560 Bq/m^3 (Gillmore et al. 2002). The authors, adopting an approximate

equilibrium factor of 0.5, estimated that the radiation exposure for a 2 h visit at the upper value of 4 MBq/m³ would be 62 mSv *per 2 h visit*.

The mining and processing of mercury ore over many centuries in Slovenia has led to a situation with significantly elevated radon activities in the town of Idrija due to various scattered mine wastes and slags. One apartment measured an indoor radon activity of between 7,300 and 15,000 Bq/m³ while outdoor radon activity averaged nearly 100 m³ in the town centre (Križman et al. 1996).

Another major ore type which may face radon issues is rare earths, commonly due to monazite but sometimes due to uranium or thorium being present also. In Australia, two important rare earth deposits at Nolan's Bore in the central Northern Territory and at Mt Weld in central Western Australia both contain radiologically significant uranium and thorium concentrations (e.g. Nolan's Bore has 18.6 Mt of ore grading 3.1% rare earth oxides, 0.021% U₃O₈ and ~0.5–0.7% ThO₂; AR 2007, 2008). Historically, monazite has been an important source of rare earths and, as noted earlier, involves significant radiological issues.

7 Brief review of health issues

As noted in the historical review, health problems in areas of high radon activities have long been known, such as the Erzgebirge. The link between radon and health impacts, however, has only been suggested since the early twentieth century—though not widely scientifically accepted until the 1960s (Jacobi 1993; NAS 1999a). By the 1930s, based on the lung cancer rates in German-Czech mines, radon was being suggested as the main cause of the health impacts, but the quantitative evidence was not sufficiently clear, especially the role of radon progeny. In the 1950s, work done by William F Bale and John Harley in the USA demonstrated that the major radiological dose was actually delivered to lung tissues by the progeny and not the radon gas (Jacobi 1993). Combined with epidemiological studies emerging by the early 1970s, it became clearer that high radon and progeny activities were associated with impacts such as lung cancers. In the same decade it also became clear that residential dwellings could also allow radon and progeny to accumulate to exposures

sometimes as high as uranium miners. Based on combined studies and evidence, the US EPA now states environmental radon as the largest cause of lung cancer in non-smokers in the USA (e.g. USEPA 2007). Radon is commonly described as the most extensively studied carcinogen (e.g. Appleton 2005).

There are two principal exposure scenarios for radon: (i) high activities and exposure with good relationships to health impacts (e.g. uranium mining), or (ii) low activities and exposures and somewhat uncertain links between exposures and health impacts (e.g. natural background radon). The evidence for radon's carcinogenic nature is derived from molecular, cellular, animal and human (epidemiological) studies accumulated over many decades (NAS 1999a). This section will present a brief review of the mechanisms for radon-induced carcinogenesis, followed by a review of health issues for high exposures and finally low exposures. Given the ongoing controversy over low radon and progeny exposures, this section is intended as a guide only. For further details, see the major studies, such as BEIR-VI (NAS 1999a) or BEIR-VII Phase 2 (NAS 2006).

7.1 Mechanisms for radon-induced carcinogenesis

The radioactive decay of radon through its progeny to a stable lead isotope involves several alpha and beta decays as well as significant gamma radiation. A single radon atom can therefore impart notable energy at the molecular or cellular level, having the potential to cause major genomic changes in a cell resulting in mutations or other transformations (NAS 1999a).

For the most common inhalation exposure situations, as noted previously, it is the progeny which is the major cause of the imparted energy (see Table 1). The issue is therefore the extent of the radon that decays while inside the lung, and the depth to which the active progeny can reach inside the lung. Finer particles can reach deeper into the lung, and hence the attached–unattached fraction is critical to consider for lung dosimetry. Radon, as a noble gas with a 3.82 day half-life, is most likely to be exhaled before decaying.

According to the most recent 'Biological Effects of Ionising Radiation' (BEIR) committee study into radon exposure and cancer ('BEIR-VI'; NAS 1999a), there is convincing evidence that most cancers are of

Table 8 Summary of global epidemiological studies for uranium or other mine-workers (BEIR VI study) (NAS 1999a)

| Cohort | Mineral | Period of follow-up | Number of miners | Person years | Average cumulative exposure (WLM) | Exposure rate (WL) | Lung cancer deaths | Age at first exposure | Year of first exposure | Length of follow-up | Excess relative risk per WLM (ERR/WLM) |
|------------------------------|-----------|---------------------|------------------|--------------|-----------------------------------|--------------------|--------------------|-----------------------|------------------------|---------------------|--|
| Yunnan, China | Tin | 1976–1987 | 13,649 | 134,842 | 286.0 | 1.7 | 936 | 18.8 | 1955.6 | 10.2 | 0.16 (0.1–0.2) |
| Bohemia, Czech Republic | Uranium | 1952–1990 | 4,320 | 102,650 | 196.8 | 2.8 | 701 | 30.1 | 1951.0 | 25.2 | 0.34 (0.2–0.6) |
| Colorado Plateau, USA | Uranium | 1950–1990 | 3,347 | 79,556 | 578.6 | 11.7 | 334 | 31.8 | 1953.0 | 26.3 | 0.42 (0.3–0.7) |
| Ontario, Canada | Uranium | 1955–1986 | 21,346 | 300,608 | 31.0 | 0.9 | 285 | 26.4 | 1963.8 | 17.8 | 0.89 (0.5–1.5) |
| Newfoundland, Canada | Fluorspar | 1950–1984 | 1,751 | 33,795 | 388.4 | 4.9 | 112 | 27.5 | 1954.1 | 23.3 | 0.76 (0.4–1.3) |
| Northern Sweden | Iron Ore | 1951–1991 | 1,294 | 32,452 | 80.6 | 0.4 | 79 | 27.4 | 1934.1 | 25.7 | 0.95 (0.1–4.1) |
| New Mexico, USA | Uranium | 1943–1985 | 3,457 | 46,800 | 110.9 | 1.6 | 68 | 28 | 1965.6 | 17 | 1.72 (0.6–6.7) |
| Beaverlodge, Canada | Uranium | 1950–1980 | 6,895 | 67,080 | 21.2 | 1.3 | 56 | 28 | 1962.6 | 14 | 2.21 (0.9–5.6) |
| Port Radium, Canada | Uranium | 1950–1980 | 1,420 | 30,454 | 243.0 | 14.9 | 39 | 27.6 | 1952.3 | 25.3 | 0.19 (0.1–0.6) |
| Radium Hill, South Australia | Uranium | 1948–1987 | 1,457 | 24,138 | 7.6 | 0.7 | 31 | 29.2 | 1956.0 | 21.9 | 5.06 (1.0–12.2) |
| Central & Western France | Uranium | 1948–1986 | 1,769 | 29,172 | 59.4 | 0.8 | 45 | 29.5 | 1956.8 | 24.7 | 0.36 (0.0–1.2) |

monoclonal origin. When combined with mechanistic processes for alpha decay and the probability of damaging cells, the BEIR-VI committee concluded that there is more evidence supporting a linear no threshold ('LNT') model for radon exposures. That is, any increase in radon exposure linearly increases the chance of cancer. The BEIR-VI committee also noted, however, that it could not exclude the possibility that a threshold dose occurred at low exposures. The BEIR-VI committee's view on the adequacy of the LNT model is supported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (see UNSCEAR 2000). The more recent and broader BEIR-VII study into low level radiation exposure re-affirmed the LNT model for radiation exposures (NAS 2006).

The model of cause-effect between radon and progeny exposures and effects such as lung cancer is at the heart of the debate about radon. A number of critical factors need to be considered in quantifying this relationship. Firstly, the combination of cigarette smoking and radon exposures is argued by BEIR-VI as synergistic, that is the combined effect of these two actions is greater than the individual sum alone. Secondly, the exposure conditions are different between uranium miners and residential dwellings, such as concentrations and equilibrium factors (exposures in miners are about one order of magnitude higher or more than indoor exposures; NAS 1999a). Finally, issues such as gender or age can also be important in the effects of exposure.

7.2 Health impacts at high exposures

The exposures of uranium miners, especially underground miners during the 1940s to late 1960s, was particularly high. The BEIR-VI committee reviewed all available epidemiological studies on uranium and non-uranium miners, with the key results given in Table 8. The combined results were used to develop the quantitative risk models in NAS (1999a), and give an indication of the 'excess relative risk' (ERR) per exposure for the various studies. The lung cancer rate relative to mean radon exposure is given in Fig. 7.

7.3 Health impacts at low exposures

The impacts of low radon exposures remain somewhat controversial. At ambient activities commonly

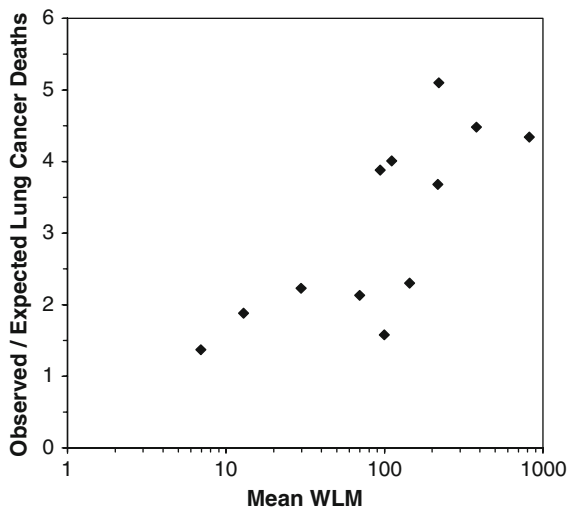


Fig. 7 Lung cancers versus radon exposure (adapted from NRPB 2000, cited by Appleton 2005)

encountered in indoor or outdoor settings, exposures are considerably lower than miners, although they can reach high levels at the lower end of typical miner exposures. Some of the major problems with many background radon versus miner studies is the different study designs, the different radon measurement techniques employed, varying radon activities encountered, variable information on confounding factors, and most importantly limited sample size (Wichmann et al. 2005). Additionally, some researchers suggest that small regions with elevated radon do not correlate to increases in lung cancers, such as radon up to 3,700 Bq/m³ at Ramsar, Iran (Mortazavia et al. 2005).

A major study which has recently begun to address the many issues associated with low exposure radon studies is the ‘Iowa Radon Lung Cancer Study’, described by Field et al. (2000). The Iowa study recently completed Phase 1 and is presently close to finishing Phase 2. At its heart is a significantly improved field methodology for monitoring and assessing the cumulative radon and progeny exposure over time, thereby reducing the major uncertainty associated with most studies to date. The Phase 1 results show that cumulative radon exposure is a contributor to lung cancer incidence (Field et al. 2000).

Similarly, an extensive compilation and combined analysis of 13 residential radon-lung cancer studies covering 9 European countries was recently published

(Darby et al. 2005). This study showed that the risk of lung cancer increased by 16% (95% confidence interval, 5–31%) per 100 Bq/m³ increase in radon after correction for random uncertainties in measuring radon concentrations. Importantly, the study again demonstrated that the LNT model was the best basis to understand exposure risk. Results from similar studies in North America also arrived at the same conclusions (Krewski et al. 2005).

8 Remediation approaches

There are two principal approaches to prevent, minimise or remediate radon problems—source reduction and dilution.

For most indoor issues, simple design and construction techniques are used to limit radon entry in the first place, and where still necessary, appropriate ventilation fans can be installed to extract ambient air and direct the radon to the external atmosphere (e.g. Lugg and Probert 1997; Groves-Kirkby et al. 2007). In any program or effort aimed at reducing indoor radon, it is critical to account for potential radon sources in building materials, as these can hamper the effectiveness of remediation designed to address underlying geologic radon sources (Groves-Kirkby et al. 2007). Some common techniques include (Lugg and Probert 1997):

- *Sub-floor depressurisation*: a wind or electric powered ventilation system is installed beneath the ground floor to extract ambient radon-rich air derived from underlying geology and eject it to the atmosphere (sumps may be included);
- *Floor sealing*: this involves placement of a low permeability material across the floor, especially focussing on filling in cracks and gaps. Given the difficulty in sealing 100% of open voids, sealing is often used in conjunction with other approaches;
- *Positive pressure*: by creating a slight positive pressure inside a building, it is possible to suppress the rate of radon entry. For this approach to be effective, the building needs to be air tight;
- *Increased ventilation*: this is essentially achieving a dilution of the radon through increased air flow through a building, however, the reductions are typically small and not sufficient for high radon scenarios;

- *Air cleaners*: given that the principal radiation exposure is derived from the reactive progeny, research has investigated this technique, although results showed only a minor reduction (the method is also more expensive than other more effective options).

For mining situations, outdoor radon is commonly considered to be sufficiently low due to atmospheric dispersion (especially diurnal processes, though atmospheric inversions may limit dispersion for brief periods).

The principal area where radon and progeny levels can accumulate very easily is in underground mining. As noted in the uranium mining section, the early years of mining uranium ores invariably involved significant to extreme radon and progeny exposures. As the link between this exposure and lung cancer incidence was more widely accepted, especially by the late 1960s, air quality standards were adopted which led to major ventilation systems being installed and significant reductions in radon and progeny exposures (e.g. Australian case study, Table 5). The design of underground mine ventilation systems to achieve desired radon and progeny levels is a complex and specialist field, as it involves fluid dynamics, mine design, particulates (especially their particle size distribution), radon exhalation, and attached–unattached progeny, as well as the interactions between aspects such as particulates and the attached progeny fraction; see papers in Gomez (1981).

Alternative research has investigated the efficacy of sealants on the walls of underground mines, such as polymers, though they are of arguable effectiveness compared to a well-engineered ventilation system (especially with respect to cost/benefits and other safety issues such as damage or fire) (see Franklin 1981). Other approaches include the use of bulkheads or backfilling to seal off sections of a mine from active operations and management, options commonly incorporated into existing or proposed underground mines. In addition, time workers spend in given areas can also be controlled to minimise exposures.

A major legacy of mining is the solid wastes remaining after mine closure. The tailings, low-grade ore stockpiles and waste rock are often locally significant radon sources, especially the tailings,

and they must all therefore be addressed during mine rehabilitation (Mudd 2008b). The traditional engineering approach is to design and construct a single or multi-layered soil cover over such solid wastes to minimise the radon flux emanating at the surface (e.g. Rum Jungle, Australia, Allen and Verhoeven 1986; Schlema-Alberoda, Germany, Schmidt and Regner 2005). There has been extensive research into the preferred soils and designs for soil covers, especially taking into account different climatic conditions (eg. sub-arctic of northern Saskatchewan in Canada versus the Rössing uranium mine in the Namib Desert of Namibia). The primary design approaches and models include the RAECOM code, developed by Rogers et al. (1984) for the US Nuclear Regulatory Commission, as well as a mixture of applied and theoretical models (e.g. Hart et al. 1986; IAEA 1992b; Ferry et al. 2001, 2002) (including the online implementation of the RAECOM code by Diehl (2008b)).

The most recent model to address radon in engineered soil covers is the coupled unsaturated flow-climate model Vadose/W (Krahn 2004), developed specifically to address the complex interactions of climatic forcing conditions (rainfall, evaporation, transpiration, temperature, wind speed, humidity, etc.), moisture flow in unsaturated (or saturated) soils as well as gaseous transport through such soils to the atmosphere. Vadose/W is intended to be used as an engineering design tool for soil covers in acid mine drainage or radon situations, and is arguably the most theoretically rigorous model yet developed for soil covers.

A common challenge is that although models are used to predict the effectiveness of different engineering designs, there appears to be very little monitoring of long-term cover performance. In Australia, the soil covers applied at the former Nabarlek, Rum Jungle, Radium Hill and Port Pirie sites have all faced ongoing problems of weeds, erosion, maintenance or poor construction (Mudd 2008b). At the Rum Jungle site, extensive acid mine drainage pollution continues despite the soil covers, though there appears to have been no studies of the performance of the covers with respect to radon fluxes (the rehabilitation target was set somewhat arbitrarily at $0.14 \text{ Bq/m}^2/\text{s}$; Mudd 2008b). The extensive remediation efforts undertaken in United States and eastern Germany, where soil covers were

constructed over substantive quantities of waste rock and tailings facilities, await a more comprehensive review and analysis—as well as the test of time.

A sound rehabilitation objective for uranium mining projects should be to return radon fluxes to pre-mining levels (Mudd 2008b). At the former Nabarlek uranium project in the Northern Territory, Australia, rehabilitation works have actually achieved an overall reduction in radon flux compared to pre-mining (Bollhöffer et al. 2006). Prior to mining, very high-grade ore (>1% U₃O₈) outcropped at the surfaced, leading to radon fluxes from 3.7 to 44.0 Bq/m²/s (Clark et al. 1981). Mining saw the tailings buried up to half the depth in the mined out pit (~100 m deep), with low grade and waste rock filling the remainder of the pit—thus leading to a weaker radon source in the near surface materials and soil covers averaging about 1.0 Bq/m²/s over the same region (Bollhöffer et al. 2006). Recently, however, a small area (0.44 ha) has shown significant erosion leading to exposure of the underlying radioactive wastes, giving a radon flux of 6.51 Bq/m²/s (Bollhöffer et al. 2006). At many projects in Australia, it appears very difficult to achieve a pre-mining radon flux after rehabilitation, especially in the long term (Mudd 2008b).

9 Conclusions

Radon is a ubiquitous natural carcinogen derived from the three primordial radionuclides of the uranium series (²³⁸U and ²³⁵U) and thorium series (²³²Th). In general, it is present at very low concentrations in the outdoor or indoor environment, but a number of scenarios can give rise to significant radiological exposures. Historically, these scenarios were not recognised, and took many centuries to understand the links between the complex behaviour of radon and progeny decay and health risks such as lung cancer. However, in concert with the rapid evolution in the related sciences of nuclear physics and radiological health in the first half of the twentieth century, a more comprehensive understanding of the links between radon, its progeny and health impacts such as lung cancer has evolved. It is clear from uranium miner studies that acute occupational exposures lead to significant increases in cancer risk, but chronic or sub-chronic exposures, such as indoor residential settings,

while suggestive of health risks, still entails various uncertainties. At present, prominent groups such as the BEIR or UNSCEAR committees argue that the ‘linear no threshold’ (LNT) model is the most appropriate model for radiation exposure management, based on their detailed review and analysis of uranium miner, residential, cellular or molecular studies. The LNT model implies that any additional or excess exposure to radon and progeny increases overall risks such as lung cancer. A variety of engineering approaches are available to address radon exposure problems. Where high radon scenarios are encountered, such as uranium mining, the most cost effective approach is well-engineered ventilation systems. For residential radon problems, various options can be assessed, including building design and passive or active ventilation systems. Overall, radon will continue to be an ever-present carcinogen and requires eternal vigilance wherever it is encountered—in mining, processing industries, building materials, caves, or even residential homes.

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