I.N.Beckman

RADON RISK

Contents

1. Introduction: Environment risk conception and radon problem

The human environment is one in which ionizing radiations are present at all times and at all places on the earth from the deepest caves to the highest mountains and on into space. Radon and its decay products are present wherever radium and thorium exist in the earth or in any planetary material. In the terrestial environment the inhaled radon isotopes and their daughter products make up almost two —third of the total dose to living tissue /1/. Radon, an inert gas, is continuously formed by the decay of its parent, Ra. Although radon itself is only a slight radiological hazard, it acts as a vehicle for dispersal of its short lived a — emmitting daughters. The frequency of lung cancer is increased by exposure to these daughters. Radon is an occupational hazard particularly in underground uranium mines and, when dispersed, contributes to the radiological exposure of the population.

Soil, rocks and most of the common building materials (except wood, glass and steel) contain small amount of naturally occurring radioisotopes of uranium and thorium series ^{222}Rn (half—life 3,82 days, called radon) and ^{220}Rn (half —life 56 s, called thoron), are gaseous radioisotopes of uranium and thorium series, respectively. Consequently, houses and other species, which contain these natural radioisotopes give rise to small amount of external gamma exposures to the inhabitants, but more important is the exposure due to radon and thoron, which can diffuse through the walls and become air — borne in enclosed spaces. Thus, gaseous radioisotopes give rise to inhalation exposures. The radiological hazard due to indoor radon results from the inhalation of the radon decay products which are short —lived isotopes of solid elements. The exposure of the lung tissue is directly correlated with the concentration of radon decay products.

A relationship has been ectablished between exposure of undegraund miners to radon daughters and induction of bronchial carcinoma. However, the exact nature of the relationship is confused by various compounding factors such as the contributing synergistic effects of smoking and/or arsenic exposure, normal metabolic variability and, perhaps most important of all, the absence of accurate radon/radon daughter exposure data /2/.

The annual effective dose equivalent per capita incurred from inhalation of radon progeny is equal to 52% of the total amount of radiation dose received from both natural and manmade radiation sources. This implies that radon and its progeny are the major contribution of natural radiation to human being. In daily activities, most people spend 80% of their time indoors, and the concentration of indoor radon is one to five times higher than that of outdoor radon. This exposure may increase even further due to new building technologies optimised for energy conversation. A relevant fraction (10 — 40%) of lung cancers in the general population are causes by the inhalation of radon. For a country such as the United States, with an average of about 40 Bq/m^3, the average

lifetime risk of lung cancer caused by exposure to radon decay products is estimated to be about 0.3%, causing on the order of 10000 cases of lung cancer annually among the U.S. population of 235 million. This average risk is more significant than that received on the average from all other natural radiation sources or from medical exposures. And the radon dose exceeds by a factor of 10 to 100 the average doses from nuclear power or weapons testing.

Moreover, in any country indoor levels a factor of 10 or more higher than the average sometimes occur. In fact, it is the common experience of the radon research community that ^{222}Rn concentrations in the range of 200 — 2000 Bq/m^3 are found with startling frequency. And although the lung cancer risk associated even with an ordinary concentration such as 40 Bq/m^3 is very large compared with many environmental insults of concern, living for prolonged periods at concentrations above 200 Bq/m^3 leads to estimated individual lifetime risks exceeding 1%. The highest values found —more than 2000 Bq/m^3 — have rises even larger than those from cigarette smoking. However, unlike smokers, those living in unusually high radon concentration are rarery aware of the large risks they may thereby be suffering/3/.

Although radon and its decay products are well known to cause lung cancer at high exposure levels, considerable controversy remains about the magnitude of risk due to low—level exposure. Linear extrapolation from the dose — response values of uranium miners who were heavily exposed to these nuclides would suggest that a relevant fraction (10 — 40%) of lung cancers in the general population are caused by the inhalation of radon daughters. Moreover, the results of monitoring programs in several countries during past years have revealed that for a small, but not negligible fraction of the population, the lifetime exposure from indoor radon daughters is comparable to, or even exceeds the occupational radon exposure of moderately exposed undeground miners still showing a significant excess lung cancer frequency.

In recent years, society's perception of the importance of indoor radon has altered drastically. Until the late 1970s, it was throught that elevated indoor radon concentrations were largely an isolated concern associated with the inadvertent presence of certain industrial and mining residues in or near buildings. Subsequent events — notably the observations around 1980 in different countries of a high incidence of ordinary houses with elevated radon levels — forced a major revision of that view within the scientific community. It is now widely understood that the most important component of radiation exposure to the public is due to the inhalation of radon decay products indoors. Even more significant is that the estimated level of health risk associated with average indoor radon levels is much higher than those due to other environmental carcinogens. Furthermore, and perhaps most importantly, radon concentrations ten or even a hundred or more times the average are observed with srartling frequency, even in buldings that are otherwise quite ordinary. Long —term exposure to these higher concentrations leads to individual risks of lung cancer that are so high as to be unacceptable by almost any standard.

Because radon mitigation tends to be dealt with more and more in multidisciplinary contexts of pollution of the indoor environment, we will outline how radon research in the future may fit into a framework often used in investigations of substances that pollute the environment. This framework is the **source—risk chain,** which consists of five stages /4/:

source—dispersion—exposure—dose—risk

The main advantage of the approach using the source —risk chain is that it shows all stages at which a problem may be controlled. For radiation protection purposes, the stochastic effects of exposure to radiation are expressed as a lifetime risk of early death by cancer or of a serious genetic effect. The risk concepts used in the assessment of non-

radioactive materials, however, are not always equivalent to the definition of "risk" used for radioactive substances. When comparing the risks of different pollutants of the indoor environment in a multidisciplinary effort, the premises for risk assessment are often discussed. It is worthwhile making an effort to bring the various approaches together.

This paper describes the application of risk methodology to solve the radon problem. The most attention will be paid efforts that should be undertaken for the reduction of man —enhanced natural radiation sources. The work concentration mainly on the radon risk estimation and classification, requirements for limiting radiation exposure due to radon and thoron, anti —radon policy, and srategies of the optimum radon risk reduction process. As a whole, the review is structured to examine the current state of knowledge of all major aspects of the radon problem.

2. Source of radon.

The mining and milling of uranium ores, uranium mill tailings, wastes arising from the treatment of other radioactive ores, for example thorium/uranium hydroxide residues arising from monazite processing, wastes, for example gypsum, from the phosphate industry since such residues contain elevated levels of ^{226}Ra, soil and rock are the source of most radon to which people are exposed. In fact, the only other sources of significance are building materials and even these generate radon because of radium that originated in the earth (or to a small degree from predecessor radioisotopes that originated in the earth themselves). In discussing radon sources, then, it is common and convenient to distinguish not only among the materials that contain the radium that leads to indoor radon, but also among the agents and pathways by which radon enters.

Despite the fact, that the sources of indoor radon are natural in most cases, the resulting exposure of the population is a consequence of human activities. Therefore, the exposure caused by indoor radon has to be considered as "man —enhanced" and not as exposure to natural sources.

2.1 The radon family.

^{238}Uranium (99.3%), ^{235}uranium (0.7%) *(at secular equilibrium the activities of the radionuclides in the uranium—235 decay chain are 4.5%* of *those in the uranium— 238 decay chain)* and ^{232}torium, the three naturally occurring actinides surving into our times in considerable amounts, are slowly transformed into the stable elements ^{206}lead, ^{207}lead and ^{208}lead, respectively. The main characteristics of these series are given in Table 1. This transformation takes place by consecutive decay chains of several unstable nuclides. With one exception, the intermediate nuclides are isotopes of solid elements. This exception is radon a noble gas, free to move by diffusion as well as convection once released from the source matrix. In the uranium series there is ^{222}Rn (radon); in the thorium series ^{220}Rn (thoron); and in the actinium series ^{219}Rn (actinon). There is no isotopeof radon in the artifically produced neptunium series. A detailed listing of the natural radioactive decay series is given in Table 2.

Radon (^{222}Rn)

Radon —222 has the longest half—life (1620 y). When radium —226 decays to radon it emits alpha particles with energies of 4.589 Mev (5.7%) and 4.77 Mev (94.3%). The alpha and radon particles recoil in opposite directions and move through the surrounding medium until their recoil energy is dissipated. The particles then become neutral atoms. Thereafter movement occurs by diffusion. Typical ranges of the alpha and radon recoil particles in various media are given in Table 3. The passage of these charged particles through solids creates narrow path of intense damage on an atomic scale. The *a* or radon particle tracks are narrow (4 — 20 nm), stable, chemically reactive centers of strain composed mainly of displaced atoms.

The radionuclides which are precursors of radon —222 in the uranium —238 decay chain are usually close to secular equilibrium in uranium ore bodies. This means the activity of the daughters can be estimated from the uraniun content of the ore. As radon an inert gas which can readily migrate, it cannot be assumed that radon —222 or its daughters are in secular equilibrium with radon —226. Many processes can disturb secular equilibrium. Examples are chemical processing, diffusion and selective sampling. In these cases, secular equilibrium will begin to be re —established once the system is "closed". The rate of approach to secular equilibrium is determined by the half—life of the daughter (or daughters) and not the parent. A useful "rule of thumb" is that secular equilibrium is approached within 5 half—lives of the longest —lived daughter. Thus radon will reach equilibrium with radium after about three weeks. Similarly, radon will be in equilibrium with its daughters polonium —218, lead —214, bismuth —214 and polonium —214 after about 3 hours. This means that if radon is measured by a — counting up to 3 counters (radon —222 and its two a —emitting daughters, polonium —218 and polonium —224) can be obtained for each radon —222 disintegration.

Although radon is only a slight radiological hazard in itself, it acts as a vehicle for dispersal of its short —lived daughters. Details of the decay chain of ^{222}Rn are given in Table 2a. There are four short—lived daughters in the ^{222}Rn series — two alpha and two beta —gamma emitters. The longest lived member is ^{214}Pb with a half—life of 26.8 m. Not shown beyond the 21-year ^{210}Pb are ^{210}Bi (5.0 d, */3)* and ^{210}Po (138 d, *a)* before this series ends at stable ^{206}Pb. From the standpoint of potential damage to the human lung the alpha decays of ^{218}Po and ^{214}Po totaling 13.7 Mev of energy are of chief concern. These nuclides are deposited in the lung through the breathing of air containing ^{218}Po, ^{214}Pb, and ^{210}Bi attached to dust or in free —ion or atomic form.

Radon is never strictly in equilibrium with its daughters as they attach themselves to aerosols which readily plate —out on surfaces. Radon daughters are also deposited on the surfaces of the respiratory tract when air containing radon is inhaled. The *a —* particles emitted when the daughters decay can result in large doses to the cells of the bronchial epithelium which lines the respiratory tract. Exposures of the lung to radon and radon decay products contribute approx. 40% of the average total effective dose equivalent of 2.4 mSv per year contracted by the general population from all radiation sources. Other natural sources of exposure to radiation include cosmic rays (12%), terrestrial gamma —rays (15%) and radioactive nuclides incorporated in the body (15%).

Of the radon isotopes, ^{222}Rn, a decay product of ^{226}Ra in soil or building materials, with its short —lived decay products constitutes the major concern as a health hazard in the environment.

Thoron (^{220}Rn).

Thoron (^{220}Rn) has a half—life of only 55 s and decays with the emission of an alpha particle having an energy of 6.29 Mev. The decay chain is given in Table 2b. The longest lived daughter is the 10.6 h ^{212}Pb which shows up in a decay series of radon and thoron decay products on atmospheric dust from a typical indoor/outdoor atmosphere. The short-lived decay series has two alphas: ^{216}Po, 6.78 MeV and ^{212}Po, 8.78 MeV (or ^{212}Bi, 6.05 MeV) comparable to the two alphas in the radon (^{222}Rn) chain. In addition there are two beta emission and some gamma rays.

It was estimated that the inhalation of short —lived decay products of radon (^{222}Rn) and thoron (^{220}Rn) accounts on average for about one half of the effective dose equivalent from all natural sources of radiation. The mean thoron progeny levels represented by equilibrium equivalent thoron concentration in dwellings seem to lie between 0.1 and 1.0 Bq m^{-3}, and so thoron progeny makes a contribution of about 10 to 30% of the effective lung dose equivalent from all indoor radon plus thoron and their progeny. However, it is reported that thoron concentration in Japanese dwellings may be

higher than in other countries because the interior walls of typical Japanese dwellings are covered with soil — based plaster. There, the dose due to indoor thoron progeny may be higher in Japan.

2.2 Origin of radon.

Uranium and thorium are widely dispersed in the environment and represent the major sources of radioactivity in the ground. Typical concentrations are 1 — 10 ppm for both uranium and thorium, in sandstone, shale or limestone and 3—12 ppm for uranium and 10 — 80 ppm for thorium in granite. The corresponding activities range from 0.02 to 0.60 Bq/g. Most construction materials contain considerable amounts of uranium and thorium. An additional source of radon may be tap water. The high mobility of the noble gas gives rise to radon water concentration which is much greater than that of its only slightly water soluble parent nuclides. Minor sources of radon are phosphate fertilizer, natural gas, coal — fired power plants and released from the uranium industry.

The ability of radon to survive migration is obviously a function of the half—life and of the permeability of source matrix. In the ^{238}U decay chain, the gaseous intermediate nuclide is ^{222}Rn with a half— life of 3.82 days. This time is short enough to restrict transport by diffusion to fairly short distances but, on the other hand, long enough to allow partial emanation from building materials and soil and to permit convective transport and migration over several meters once the radon has reached air or water spaces. The isotope of radon produced in the ^{235}U decay chain, ^{219}Rn (actinon) has a half—life of only 3.96 s. The migration of this isotope will be restricted to a few millimeters and practically no ^{219}Rn will reach the surface. In the ^{232}Th decay chain, one intermediate nuclide is ^{220}Rn (thoron) with a half-life of 55.6 s. Due to the short half—life only a small fraction of thoron can emanate from building materials or infiltrate from the ground since migration is restricted to a few centimeters. With the exception of thorium —rich areas and thorium mining, the concentrations of ^{219}Rn and ^{220}Rn in air are negligible compared to those of ^{222}Rn.

Uranium

Uranium is widely distributed throughout the earth's crust to extent of about three parts per million. Minerals of commercial value containing several thousand parts per million (ppm) of uranium are ordinarily oxides such as uranite and carnotite with phosphates and monazite sands being of importance in some cases. Rich ores will contain from 1% to 4% uranium.

It should be noted that phosphate rocks widely used as agricultural fertizers contain quantities of uranium of radon of from 8 to 400 ppm with a mean value of about 40 ppm. In general ^{226}Ra is in equilibrium with the ^{238}U; however, an appreciable fraction of the ^{226}Ra is removed as a byproduct in preparation of the fertilized. The dose received by the general population from this source is not considered significant.

Thorium

Thorium ranks just under lead in abundance in the crust at 11 ppm. It is obtained chiefly from monazite sands, which contain approximately 10% thorium providing high external dose rates for residents of coastal areas where they are found. It is used in magnesium — thorium alloys in aircraft and space vehicles in addition to gas light mantles and for certain phosphors in welding. ^{232}Th exceed ^{238}U by a factor of from 3 to 5 in many rock types. Granite rocks exceed basalts in the igneous category and shales are more productive than carbonates (e.g. limestone's) in the sedimentary class. As noted in the table the crustal abundance of ^{238}U is estimated to be 2.8 ppm (36 Bq/kg) while ^{232}Th is present in soil and rock at about 10.7 ppm (44 Bq/kg). Hence, although ^{232}Th exceeds ^{238}U in the continental crust by a factor of 3.8, the activities are almost equal. ^{232}Th activity exceeds of ^{238}U by only about 20%.

Radium

The ^{226}Ra, ^{224}Ra and ^{226}Ra distributed in crustal soil and rocks release isotopes of the inert gas radon to capillare and pores where it can reach levels of the order of 1000 times that of ordinary outdoor air. The result is a steady transport of radon from soil to air.

Crustal rocks and soils contain about 40 Bq/kg of ^{226}Ra with granites generally exceeding limestone and sandstones by a factor of three or so. Building materials reflect concentrations of their crustal sources with concrete ranging up to 70 Bq/kg and bricks even higher. The chemical behavior of radium in soils is sufficiently different from uranium to the extent that individual samples may show clear departures from equilibrium in the uranium series. This provides ample time for chemical changes and redistribution of elements to occur in near —surface soils. Radium in the form of chlorides is found in water supplies from lakes or streams to the extent of 0.5 to 50 Bq/m^3. Water drawn from wells will generally have a higher ^{226}Ra concentration than those from surface supplies. Sea water contains a relatively low concentration of about 4 Bq/m^3.

The radium extensively used for medical purposed for many years, and the parent of the ubiquitous radon which has received wide notoriety in recent years as a possible danger in homes and work places.

2. **Dispersion of radon**.

The effectiveness of radium in supplying radon to the soil pores for transport to the atmosphere depends not only upon the total concentration of radium atoms present per unit mass but also upon the fraction of those atoms in the soil and rock which are located in the matrix or on soil particle surfaces so that the newly formed radon atoms can escape into the pores and capillaries. The ratio of the radium atoms so situated to the total number of radium atoms present is called the "emanation coefficient". This fraction is also referred to as "emanation power" or "emanating fraction" in the literature. Values vary widely from 0.02 in lava fields in Hawaii to 0.76 in agricultural soils nearby. The concept of "emanation coefficient" can best be illustrated by the surface —to—volume ratio for a given set of particles making up a soil. The diffusion coefficient for radon atoms in a solid is only about 10~^{7} of that for air. Hence, only atoms formed very near or on the surface of soil particles will reach the soil air space.

The transfer of radon atoms from mineral grains to soil capillaries is also dependent upon water films in the interstitial pore spaces. The recoil range of a radon atom at the instant of decay of the parent radium atom is about 69 */um* in air, 0.006 */u.m* in water, end even less within a mineral grain. If the average distance between soil particles is less than the recoil range of the radon atom in air, many of these recoiling atoms (or ions) will penetrate the surface of the neighboring soil grain and be lost to the soil pore space. On the hang, if a film of water surrounds the grain, there is a higher probability that it will remain in the air space between soil particles, If too much water is present, soil transport procedures are impeded since the diffusion coefficient in water is of the order of 10 ~^{3} of its air value.

The a mean emanation coefficient of 0.36 were obtained for soil samples from New Mexico, Texas, Hawaii, Alaska and Iceland. Even though thorium is generally more abudant than uranium in the earth's crust, the probability for decay is smaller; hence, the production rates of ^{222}Rn and ^{220}Rn in the soil are roughly the same. Much of the thoron decays before reaching the earth's surface due to its short half-life of 52 s.

There are several characteristics of radon and thoron that account for the dominance of these isotopes, and their short —lived daughters in the dose to the human respiratory tract. These factors include (1) the spread availability of the parent nuclides, ^{226}Ra and ^{224}Ra, in the rocks and soil and building materials, (2) the transport processes available for radon as an inert gas, (3) the significant list of short —lived daughter products, and (4) an alpha

energy in each series of approximately 25 Mev per decaying radon or thoron atom. The result is that inhaled radon progeny account for 55% of the annual dose to the human compare d with nearly equal doses of about 15% each from the other natural ionizing radiations including cosmic rays, terrestrial radiation and natural radionuclides such as ^{40}K in the body.

3.1 Outdoor radon 3.1.1 Radon migration

Radon transport processes below the earth's surface — Radon migration in the ground.
—Migration of radon isotopes into rock pores.
—Diffusion of radon gas into soil cavities.
—Radon in mines
—Radon emanation from uranium ore
—Radon exhalation from mill tailings
—Radon in natural gas and petroleum
—Radon exhalation from soil.
—Radon releases from uranium mill tailings (bare and covered tailings)
—Emanation of Rn from coal ash
—Desorption of radon at the Earth's surface
— Episodic radon changes in subsurface soil gas along active faults and possible relation to earthquakes.
3.1.2   Geological approach to radon problematic.
—   regional radon risk maps
3.1.3   Geographic and site characterization of radon source potential
—   radon level mapping (radon emission risk)
3.1.4   Radon in the atmosphere
3.2 Radon and its decay products in indoor air.
3.2.1   Generation in migration indoor radon
—Indoor concentrations
—Soil as a source of indoor radon: generation, migration and entry
   —Building materials as a source of indoor radon (natural radionuclides in building material, exhalation of radon from building material)
—Radon migration through walls
—Radon entry via potable water.
— .The flow of radon through cracks in a concrete slab
3.2.2   Regional radon risk maps
—   radon potential mapping
— maps of radon risk at the building site (new and old houses)

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