Chapter One Indoor Radiation – A Measurement of Environmental Radon and Thoron Concentrations in a Multi-Storey Apartment Building

1.1 Introduction

Natural radiation has always been a part of the human environment. Its main components include cosmic radiation, terrestrial gamma radiation from natural radionuclides in rocks and soil.

Radiation is defined as energetic particles or radiation generated by nuclear reactions. The main sources of environmental radiation are attributable to cosmic rays (galactic and solar in origin), in which the radiation will come from neutrons, protons, electrons and photons. Radioactivity in the natural environment of earth's atmosphere here, on the other hand, leads to radiation of the alpha and beta particles, gamma rays, and neutrons. Natural environment radioactivity would also be about 80% of man's radiation exposure on earth. Alpha particle releases from atmospheric radon are the most important due to its nature of discrete energy (Smith, F.A., 2000).

Unstable nucleus becomes more stable by emitting a particle in the process known as radioactive decay.

Alpha decay - Alpha particle emission consists of 2 protons and 2 neutrons. The emissions from alpha decay are discrete in energy.

Beta decay - Beta decay takes three forms, they are Beta$^-$ emission, Beta$^+$ emission, and electron capture. Being isobaric decay, mass number of the nucleus does not change.

The present investigation aims at the detailed study of indoor radiation environment, which is in terms of the alpha activity concentration, radon and thoron concentration levels and the inhalation dose resulting from these gases and their progenies. Radiation dose imparted by inhalation of indoor radon contributes more than 50% of the total radiation dose. Indoor radioactivity in the environment therefore is a matter of health concern. Furthermore, a higher upper value of radiation level used to be tolerable but it has been revised downwards in 2007.

> *"Taking account of the new findings, the Commission has therefore revised the upper value for the reference level for radon gas in dwellings from the value in the 2007 Recommendations of 600 Bqm^{-3} to 300 Bqm^{-3}." (ICRP, 2007)*

In this thesis, a study on the indoor radon and thoron concentrations in a multi storey apartment is presented. A residential apartment block of fifteen stories has been chosen and radon and thoron concentrations were monitored in different units in the different floors.

1.2 Literature of Indoor Radon and Thoron Measurements in Dwellings around the World

Around the world indoor radon and thoron have been measured to account for human exposure. Radon inhalation especially is associated with lung cancer risk. Varying indoor radon works have been carried out in different types of regions, old and new setting, different building types, different building functions, varying seasons impact on indoor radon also have been taken into account in the climatic zones. There has also been a mixture of both to compare for ratios, before advance building technology remediation, all for the case of indoor dwelling measurements.

In the countryside of Xinwen and Dukou, China, equilibrium equivalent thoron concentrations measured in two dwelling types of red brick and soil wall were: 1.15 Bqm^{-3} (red brick wall) and 5.26 Bqm^{-3} (soil wall) in Xinwen, 0.61 Bqm^{-3} (red brick wall) and 3.68 Bqm^{-3} (soil wall) in Dukou, respectively (Zhao et al., 2008).

In Chhattisgarh, India, the geometric mean of indoor thoron concentrations in the urban dwellings have been measured to vary from 11.57 to 25.88 Bqm^{-3} , its overall geometric mean value is 16.65 Bqm^{-3}. While in rural dwellings, measurements vary from 12.50 to 30.08 Bqm^{-3} with an overall geometric mean value of 19.00 Bqm^{-3}. The indoor radon concentrations in the urban dwellings have been measured to vary from 20.20 to 30.31 Bqm^{-3}, while in rural dwellings it measures from 15.50 to 36.05 Bqm^{-3}. The overall geometric mean of the PAEC levels in urban dwellings are 2.58 and 1.50mWL for thoron and radon respectively. In the rural the values are 4.57mWL for thoron, and 1.87mWL for radon. Chhattisgarh's annual geometric mean value of inhalation dose is found to vary from 0.39 to 1.36 mSv y^{-1} , in the urban dwellings, and its overall mean value is 0.60 mSv y^{-1}. In its rural area the annual inhalation dose is varying from 0.39 to 1.46 mSv y^{-1} , with a geometric mean value of 0.74 mSv y^{-1} (Khokhar et al., 2008).

In Ajloun, Jordan , the villages under investigation measured the highest radon concentration in the dwellings made of clay (45.7± 6.7 Bqm^{-3}), and the lowest was in dwellings made of brick (33.9±6.4 Bqm^{-3}). The average indoor radon concentration was 36.3±2.3 Bq m^{-3} for the measurements of the whole survey and that corresponds to an annual effective dose of 0.92±0.06 mSv y^{-1} (Al-Khateeb et al., 2012).

In the new region of Adapazari, Turkey, average radon concentration (arithmetic mean) measured a minimum value of 24.0 Bqm^{-3}, a maximum value of 108.5 Bqm^{-3}, with a mean of 63.5 Bqm^{-3}. In the old region of Adapazari, on the other hand, dwelling average radon concentration measured a minimum 16.2 Bqm^{-3}, a maximum 155.0 Bqm^{-3}, with a mean of 59.9 Bqm^{-3} (Kapdan and Altinsoy, 2012).

In the case of UK dwellings, the mean radon level is approximately 20 Bqm^{-3}, compared to 4 Bqm^{-3} in the outside air (Wrixon et al., 1988). Levels up to 17,000 Bqm^{-3} have been measured in residential properties in the West of England (National Radiological Protection Board, 2004). In a 5-week study before engineering remediation, indoor radon levels monitored in a two-storey detached dwelling in Northamptonshire, the upstairs measured 299.06 Bqm^{-3} (95% C.I. 45.58), the downstairs measured 370.14 Bqm^{-3} (95% C.I. 75.98). As part of this project study, sub-slab depressurisation remediation measures were installed at halfway point (Allison et al., 2008).

Presently the Malaysian scenario is on an uptrend in high-rise buildings being constructed in the urban city. With persisting intense area infrastructure upgrade, soil excavation works are causing continued exposure to radon sources too.

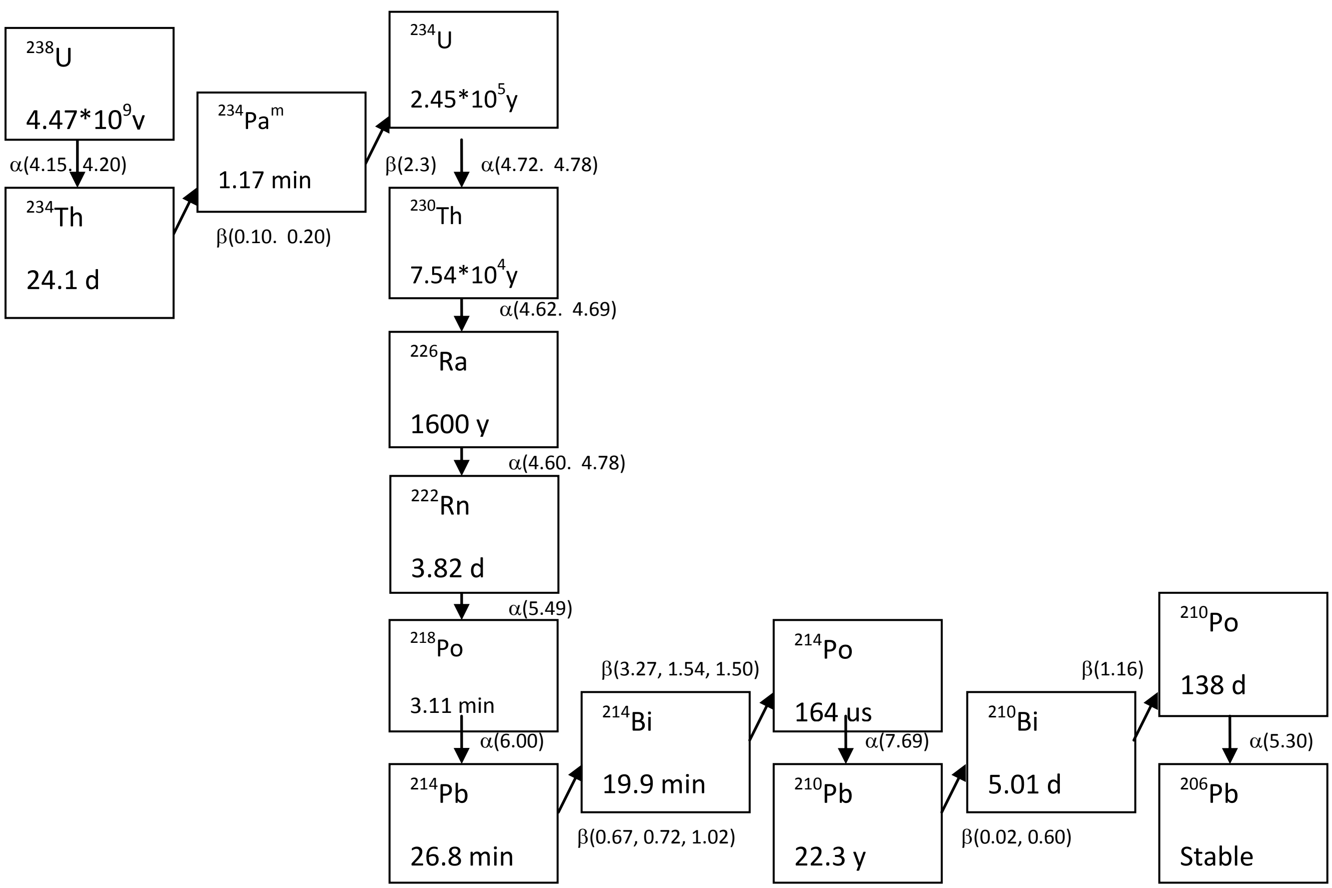
1.3 The Radioactive Series

Naturally occurring radioactive materials (NORMs) are broadly distributed throughout the environment. NORMs on earth exist because of their long half-lives, and their constant production by natural processes, such as the interactions of cosmic rays with atmosphere, and their surfacing as decay products of long-lives radionuclides. These radioactive nuclides occurring naturally have atomic numbers greater than that of lead, and each of these can be arranged in one or other of three series. The radioactive series are namely the uranium series (ends with Pb-206), the thorium series (ends with Pb-208), and the actinium series (ends with Pb-207). The final member of each series is a stable isotope of lead with a different isotope in each case. Many rocks of the formation strata where oil and gas reservoirs are found contain NORMs.

1.3.1 The Uranium Series - Radon and its Progeny

Referring to Figure 1.1, in the uranium-238 series, radon-222 is a decay product in this series, which then alpha decays in 3.82 days to become polonium-218. Polonium-218 takes three minutes of alpha decay to become lead-214 and then another 27 minutes of beta decay to become bismuth-214. Airborne concentrations of polonium-218, lead-214, and bismuth-214 are of radiological significance owing to their potential retention in the lungs and subsequent irradiation by the alpha decays of these radionuclides. These radioisotopes are radon progenies with short half-lives accounting for relatively high inhalation dose to the lungs. They are decaying in the lung before potential for clearance by i) absorption into blood, or by ii) particle transport to the digestive gastrointestinal tract (Marsh et al., 2008).

Figure 1.1: Decay Chain of Uranium-238 Including Radon-222 and its Decay Products
Decay Chain of Radon-220 (Nero Jr., p.6, 1988)



1.3.2 The Thorium Series - Thoron and its Progeny

Thoron is a nuclide product in the thorium decay chain. It originates from the primordial thorium-232 of 14 billion years ago (refer Figure 1.2). Thoron decays emit alpha particle with a half-life of 55.6 s. The decay products of thoron emit alpha and beta particles with half-lives in the range between 310 nanoseconds and 10.6 hour (Meisenberg and Tschiersch, 2010).

There is relatively little information on the characteristics of the thoron progeny aerosol in room air and the degree of radioactive equilibrium. However, in Ramola et al. (2012),

> *"Thoron decays through the short-lived daughters ^{216}Po, ^{212}Pb, ^{212}Bi, ^{212}Po and ^{208}Tl to the stable nuclide ^{208}Pb. The lung and other organ doses contributed from ^{220}Rn and its progeny were usually neglected in the dose assessment because of generally low concentrations compared with the ones of ^{222}Rn progeny. On the other hand, as the half-life of thoron progeny (^{212}Pb, 10.64 h) is much longer than that of ^{222}Rn progeny, and the alpha energy emitted from ^{220}Rn progeny is high (^{212}Po, 8.78 MeV), the effective dose per unit equilibrium equivalent concentration of ^{220}Rn (EEC_{Tn}) is more than four times higher than that of equilibrium equivalent concentrations of Rn-222." (Ramola et al., 2012)*

In general, therefore, more than 90% of the potential alpha energy associated with thoron progeny in indoor air is carried by Pb-212 (Nero Jr., 1988).

Figure 1.2: Decay Chain of Thorium-232 Including Radon-220 and its Decay Products (adapted from Nero Jr., p.44, 1988)

Th-232	→	Ra-228	→	Ac-228	→	Th-228	→	Ra-224	→	Rn-220
(1.4×10^{10} yr)		(5.8 yr)		(6.1 hr)		(1.9 yr)		(3.6 d)		(55 s)
										↓
Pb-208	←	Po-212	←	Bi-212	←	Pb-212	←	Po-216		
(stable)		(0.3 s)		(61 m)		(10.6 h)		(0.15 s)		
				↓						
				Tl-208						
				(3 m)						
				↓						
				Pb-208						
				(stable)						

1.4 Attachment of Radon Progeny to Aerosol Particles

Air may contain a mixture of solid or liquid particles known as aerosols. They range in sizes. Cooking and smoking are prominent sources of indoor aerosols. Aerosols play an important role in the behavior of radon progeny indoors. Radon and its progenies can leave the indoor by exfiltration and also by decay. The progenies can also attach themselves to aerosol particles. Radon and radon progenies, and thoron and thoron progenies, are solids that attached to aerosols and inhaled into lungs. Whether attached or unattached, both radon and thoron tend to deposit on and stick to surfaces exposed to the air (Knutson, 1988).

1.5 Sources of Indoor Radon

According to Nazaroff et al. (1988), **s**ubsoil sources are the only cause and supply of radon. Soil and bedrock on earth contains primordial radionuclides of uranium, thorium and potassium all through the globe albeit in varying amounts. Radon enters into indoor atmosphere via pressure-driven flow of gas in soil through fine openings (cracks, small holes) in the floor. Building air pressure is normally lower inside than the air outdoors. The under-pressure is due to the air being warmer in the inside compared to the outside. Lastly, effects of pressure differences, wind blowing indoor, relative humidity and soil moisture will contribute to drawing radon into a building.

Figure 1.3 portrays a subsoil ground and explains for the generation of radon and its movements. Bottom of figure firstly attribute radium decay as a source to radon availability. The characteristics of soil then influence the rate of emanation of radon into the pore air of the soil. According to Figure 1.1, two parameters to consider are the radium content and the emanation coefficient. Some radon remained in pore spaces, some radon migrate through molecular diffusion. From one state to another, radon continues to decay, some become released into the atmosphere, some radon find entry into a building. Lastly, the actual entry rates of subsoil radon into a structure are influenced by the characteristics of the building shell and the building operation (Nazaroff et al, 1988a).

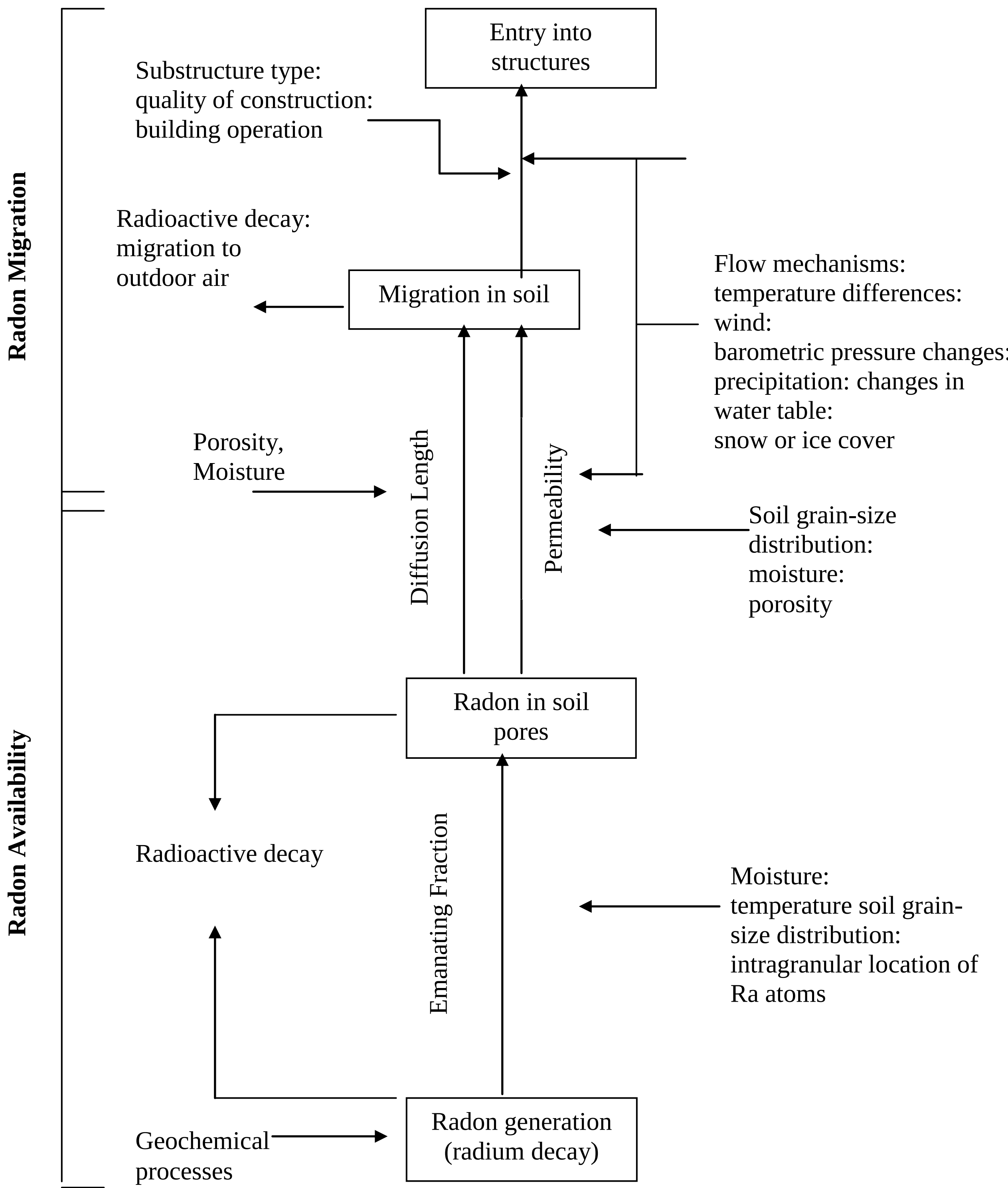


Figure 1.3: Schematic representation of radon production and migration in soil and its entry into building. (adapted from Nazaroff et al., p.59, 1998a)

1.6 Radon in Building Materials

Even though many building materials produce radon, there are certain materials that produce more radon contributing significant sources of indoor radon. Such materials have a combination of elevated levels of ^{226}Ra (the radioactive parent of radon) and a porosity that allows the radon gas to escape. Examples are Italian tuff, lightweight concrete with alum shale, and phosphogypsum.

From experimental data in a selection of building materials (Faheem and Matiullah, 2008, Stoulos et al., 2003), natural radioactivity occurs in building materials. Low and varying amount of radon exhalation properties of different building materials subsist. However, the actual use of the materials varies in different homes. For instance, concrete is used as the main building material in blocks of apartments. Ceramic tiles, on the other hand, are used in only small quantities in a dwelling. The importance of building material will thus depend not only on the specific exhalation rate of the material, but also on the actual use of the material. Some building materials, such as alum shale concrete in Sweden, have very high radium concentration and radon exhalation rates. Even though stone materials in thermal magazines studied by Stranden and Nyblom were "normal," the stone magazines contributed significantly to the indoor radon concentration in several of the houses investigated. This is due to the fact that the mass of stones is large in such magazines, and the radon is transported out of the stone magazine by the flow of air. However, building materials usually contribute relatively little compared with soil to the indoor radon concentration (Stranden, 1988).

1.7 Other Factors of Indoor Radon Accumulation

Apart from the availability and diffusion of radon into a substructure being sources of concentrations of indoor radon, building ventilation and building characteristics are influencing the variation in the concentration of indoor radon.

Firstly, characteristics of the building in aspects of design and mechanism for ventilation may affect the pressures driving bulk airflow through soil. Secondly, characteristics in the design and construction of the substructure will affect the movement of air in the soil and air in the building (Nazaroff, Moed, Sextro, 1988).

Ventilation refers to the total supply rate of outdoor air into a building. Intermittent activities of building occupants may influence the ventilation, the indoor-outdoor pressure differences, and the subsequent indoor radon concentration. In terms of the importance of ventilation, ventilation helps with controlling odours, maintaining a proper balance of metabolic gases, and diluting pollutants generated by indoor sources.

The most important aspect of ventilation for radon entry from soil is its relationship to the pressure difference between the base of the outside walls and the building's substructure openings. In an unbalanced mechanical ventilation system, when ventilation is provided in excess, pressure inside a building is increased, and air flows out either through designed penetrations or through leaks in the building shell. This in principle is a beneficial side effect of reducing radon concentration (Nazaroff et al., 1988a).

1.8 Objectives of Study

The study assessed for the space variation of indoor radiation in a multi-storey apartment building in Kuala Lumpur. The objectives of the study are as follows:

a) To measure Potential Alpha Energy Concentration (PAEC), radon and thoron concentration.

b) To study the annual effective dose equivalent (inhalation dose) in a multi-storey apartment building.

c) To investigate the variation of PAEC, radon and thoron concentrations as a function of floor heights (using floor level as a proxy).

d) To investigate the variation of radon and thoron concentrations as a function of floor furnishings.

1.9 Organisation of the Thesis

This thesis is presented in five chapters.

An introductory overview of the natural environmental radiation, radium, and thorium decay series, their sources are presented in this first chapter.

The second chapter is devoted to a review of the lung dosimetry and health effects of radon. The experimental techniques employed in this study are presented in the third chapter. The results obtained are detailed and discussed in the fourth chapter. Finally the conclusions drawn for this study is presented in the fifth chapter.

Chapter Two A Review on Dosimetry and Radiological Risks

2.1 Radiological Importance of Radon

The ultimate reason for all works on indoor radon is the potential for exposures to radon decay products to cause ill effects among humans. A basis for this concern has been the increased incidence of lung cancer among mine workers exposed to higher than average levels of ^{222}Rn decay products. This led to some estimation of the risk of lung cancer from indoor exposures and of the importance of this radiological risk relative to other environmental insults.

Inhalation dose imparted by radon and its short lived progeny can be as high as 59% of the total dose in the natural background radiation areas. It is a health concern especially true for regions where concentration of radon is high, or possible accumulation of radon and its progeny is high.

World average of annual effective dose to a human by natural radiation is 2.4 mSv and about half of which is due to indoor radioactivity, and indoor structures are places of long-term human exposure to high concentrations of radon, thoron and their progenies. There are two types of biological effects due to radiation exposure: stochastic effect and deterministic effect. In stochastic effect, the chance of the effect occurring, is statistical in nature and is a function of dose, without any threshold. Examples are cancer and genetic effects. For deterministic effects, groups of cells or tissues are extensively damaged due to exposure to very high radiation doses and the biological effects may appear, within a few hours to a few weeks, after exposure to ionizing radiation. The effect has a threshold and the severity of the effect, is proportional to the dose received. Reddening of the skin is an example of such effect.

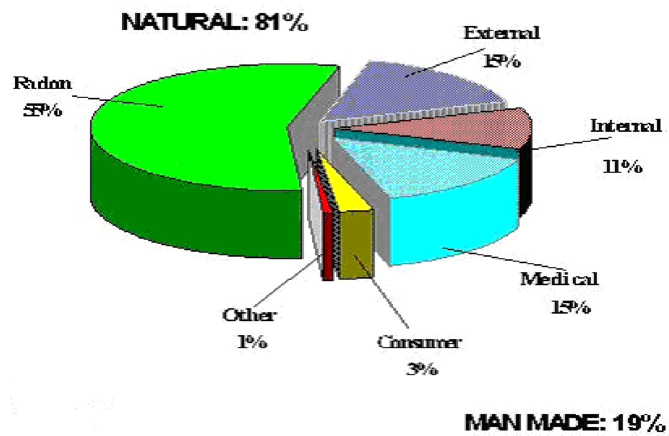


Figure 2.1 Contribution of radiation dose from different sources (UNSCEAR, 1993)

2.2 Health Risks of Radon

Referring to Figure 2.1, man-made sources constitute nineteen percent of radiation dose. These categories may be intense radiation dose as they are applied for medical and consumer use (industrial and nuclear fuel recycle). Natural radiation sources, on the other hand, constitute eighty-one percent to our radiation dose. Under this category, radon is estimated to cause a maximum radiation dose of fifty-five percent. Furthermore, coal mining, any other mining, above ground daily workplaces; these activities too, are creating by-products that become natural sources of radiation (UNSCEAR, 2000).

Radon gas is chemically inert and does not react with body tissues. While some inhaled radon dissolves in the body fluids, the resulting concentration is low and the radiation dose from the radon gas itself is negligible. Radon progenies on the other hand are solid particles. Most of the radon progenies become attached to tiny dust particles (i.e. aerosols) in the indoor air, while a variable volume remains unattached. In Figure 2.2, inhaled particles containing fractions of both attached and unattached radon progenies are deposited in the lungs. Alpha particles emitted by radon progenies inside the lungs are absorbed by nearby lung tissues. The penetration range of alpha particles is not more than a fraction of a millimetre, thus only the immediate area of the tissue will be affected and endure damage. Other radon progenies emit beta particles and gamma rays too which will have longer travel range inside the lungs.

If radon gas is inhaled, these radioactive nuclides may become attached to the inner linings of the lungs. Depending whether radon are attached to particles or not, they will be lodged in the respiratory tract and continue to decay by emitting alpha radiation. These processes damage cells in the lung tissue by creating free radicals or causing DNA breaks, and mutations that turn cancerous.

The extent of risk of developing lung cancer depends on the radon concentration in the air and the duration of exposure. Other factors such as smoking, exposure to hydrocarbon fuels may increase the risk of lung cancer. Smokers exposed to radon progenies are at greater risk of developing lung cancer.

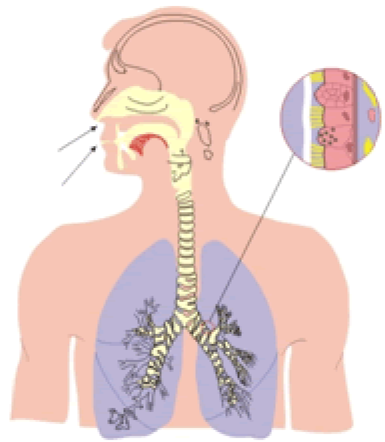


Figure 2.2 Radon gas enters the lungs, which increases the health risk (USEPA, 1992)

2.3 Smoking, Radon Inhalation, and Lung Cancer

Lung cancer is synergistic with smoking and radon exposure. The combined effects leading to lung cancer risks are exceeding the sum of their independent effects. This arises because the progenies of radon often become attached to smoke and dust particles as inhaled aerosols, and are then lodged in the lungs.

2.4 The Respiratory Tract

The respiratory tract is divided and viewed as three regions based on anatomy and function (refer Figure 2.2). The airflow and structure in each region determine the deposition of inhaled aerosol particles that may include any unattached radon progeny. For radiation dose absorbed, thickness of the epithelium may be influencing in the sensitivity of absorption.

The nasopharyngeal region starts at the nostrils down the respiratory airway to the larynx. Nostrils or the nose primarily humidify air that we breathe and protect the respiratory tract by filtering out dust and aerosol particles.

Nasal deposition of radon progeny potentially reduces the dose to lower respiratory tract. Though there is no evidence from epidemiologic mining population studies that risks of nasal cancer have increased from exposure to radon progeny. The main risk is always that of bronchogenic lung cancer (Nazaroff et al., 1988b).

The tracheobronchial region starts at the larynx, consists the trachea and the bronchial airways, down to the terminal bronchioles. Then there are about ten branchings of large airways known as bronchi, followed by about five generations of bronchioles. The bronchioles further branch into alveolated airways, where gas exchange occurs in the lungs. The tracheobronchial region is ciliated with mucous-secreting glands and so deposited particles can be cleared by mucociliary action down the throat. These are the pathways of travel in the respiratory tract exposed to radon and progenies irradiation.

The alveolar-interstitial part of the lungs is the alveolar ducts, terminal sacs, the blood capillaries, and lymphatic drainage ducts. For alpha particles of 50-80 microns, the alveolar-interstitial region is almost homogeneous. And all types of cells here absorb similar doses if affected by radiation.

Particles still intact are cleared from the alveolar-interstitial by two routes: in macrophages to the ciliated airways, and lymphatic drainage ducts finally to the regional lymph nodes. Here is the stage for lung clearance of any residue and translocation of radon progeny activity to other organs, and absorption into the blood, if any, also occurs here. Though in this region, short-lived radon progeny do not pose any importance of risk in these pathways.

2.5 Lung Dosimetry and Lung Cancer

By estimating the radiation dose absorbed and its distribution to sensitive respiratory cells, health risks caused by indoor radon and its progenies in causing lung cancer can be established. This procedure has been established for radon progeny and other inhaled radionuclides by the International Commission on Radiological Protection (ICRP). The composite of the quantity in reference related by the ICRP to risk of radiation dose indicates the effective dose equivalent as mentioned by Nazaroff et al. (1988b).

Tissue thickness in relation to the alpha-particles ranges is a determining factor since alpha particles are short range in radiation, and so dose absorbed is only affected by the sensitive epithelial cells, which are given in Table 2.1 (James, 1988).

Table 2.1 Ranges in Tissue of Alpha Particles Emitted by Radon and Thoron Progeny
(Table adapted from James, 1988)

Nuclide	Energy (MeV)	Range (µm)
Rn-222 (radon)	5.49	41
Po-218 (RaA)	6.00	48
Po-214 (RaC’)	7.69	71
Rn-220 (thoron)	6.29	52
Po-216 (ThA)	6.78	58
Bi-212 (ThC)	6.06 (36%)	49
Po-212 (ThC’)	8.78 (64%)	89

As shown in Table 2.1, the progeny of thoron Po-212 carries the most energy for penetration in sensitive lung tissue and penetrates the furthest range within the lungs.

Table 2.2: Distribution of Tumour Types in 28,000 Cases of Lung Cancer
(Percy and Sobin, 1983)

Tumour type	Frequency (%)
Squamous cell carcinoma (epidermoid tumour)	32
Adenocarcinoma	26.5
Small cell carcinoma	16.5
Large cell carcinoma	7.5

According to the Table 2.2, the epidermoid tumour, or squamous cell carcinoma tumour is the most commonly diagnosed tumour type in cases of lung cancer.

Earlier studies of uranium mining population lung cancer health problems at the Colorado Plateau of the United States revealed that about two-thirds of the cases were small cell carcinoma tumours lung cancer. Subsequent follow-up of the remaining mining population who were alived from 1954 through 1979, revealed another 292 cases of lung cancer of the adenocarcinoma tumour. The incidence of tumour types among miners is not significantly different from that in the general population (Nazaroff et al., 1988b).

Today, the EPA of the United States (USEPA, 2012) has estimated that 21,000 annual lung cancer deaths are radon-related, it is a factually established second leading cause of lung cancer:

> "***Radon*** *is the number one cause of lung cancer among non-smokers, according to EPA estimates. Overall, radon is the second leading cause of lung cancer. Radon is responsible for about 21,000 lung cancer deaths every year. About 2,900 of these deaths occur among people who have never smoked*. (USEPA, 2012)."

2.6 Other Effects of Exposure to Low Levels of Ionising Radiation

It is unknown whether other sources of radioactive decay, or radon in the background natural radiation, causes other types of cancer such as leukemia. It is indeterminate with cancer cases if persons working in or living near radioactive facilities, and persons subsequently exposed to radiation may have arisen due to chance, or unlikely due to chance, in observed excess cancer cases in Britain (BEIR V, 1990). Various studies have been made to assess the relationship between radon and leukemia, and these epidemiologic studies diverge between residential radon exposure and miners' radon exposure (Law et al., 2000).

Inhaled radon does become excreted by exhalation before significant radiation dose to the lung tissue is deposited. Ninety percent (90%) of absorbed radon through ingestion of dissolved radon in drinking water will also be excreted by exhalation within 100 minutes (US Public Health Service and EPA, 1990). Acknowledging health hazards that may arise from radon and its progenies, assistance for tests and assessments on our indoor environment can be a continuous effort.