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Murad AL-MUGAHED

RADIOLOGICAL RISK ASSESSMENT OF RADON EXPOSURE: ANNUAL EFFECTIVE DOSE AND EXHALATION RATES FROM BUILDING MATERIALS, SOIL AND FERTILIZERS USED IN YEMEN AND MOROCCO

JURY

El Hassan SAIDI,	PES, Faculté des Sciences, Université Mohammed V– Rabat	Président
Abdelkader BOULEZHAR,	PES, Faculté des Sciences, Université Hassan II, Ain, Chock Casablanca,	Rapporteur / Examinateur
Mohammed CHERRAJ,	PH, Faculté des Sciences, Université Mohammed V– Rabat	Rapporteur / Examinateur
Farida BENTAYEB,	PES, Faculté des Sciences, Université Mohammed V– Rabat	Directeur de thèse
Rachid AHL LAAMARA,	PH, Faculté des Sciences, Université Mohammed V– Rabat	Rapporteur / Examinateur

Année Universitaire : 2019-2020

✉ Faculté des Sciences, avenue Ibn Battouta, BP. 1014 RP, Rabat –Maroc
☎+212 (05) 37 77 18 34/35/38 🖷 +212 (05) 37 77 42 61 ; http://www.fsr.um5.ac.ma

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Abstract

This research work aims to evaluate the annual effective dose expected to be received by populations due to exposure to radon from 68 different samples of the most widely used traditional building materials in Yemen and Morocco. In addition, we have evaluated radon concentrations in 127 points in the Qa'a Al-Hakel soil in Yemen and 12 samples from fertilizers used in agriculture in the same area.

The closed-can technique has been employed in this study using solid-state nuclear track detectors (CR-39).

The local and imported samples were collected from various building materials that are available in shops, quarries, factories and homes under construction.

Results obtained from the current study show that the annual effective dose in all samples varies from 18.03 to 127.77 μSv y^{-1} with an average value of 55.74 μSv $y^{-1}$for building materials in Yemen and from 3.82 to 155.83 μSv $y^{-1}$with an average value of 27.35 μSv y^{-1} for building materials in Morocco. The annual effective dose for each sample was also determined and compared with the effective dose limit values recommended by the National Council on Radiation Protection and Measurements from 1 to 10 mSv y^{-1}.

In general, the annual effective doses from the investigated building materials are low and fall under the average global value.

The results show that Qa'a Al-Hakel soil contains high radon concentrations due to NPK fertilizers being used in agriculture and containing high concentrations of radon gas. The use of these fertilizers leads to the accumulation of radionuclides in soil, which can lead to serious health risks.

The effective radium content was calculated as well. All the values of effective radium content in all samples tested were found to be quite lower than the permissible value.

Keywords: Effective dose, radon exhalation; CR-39; building materials

Résumé

Ce travail de recherche a pour objectif d'évaluer la dose efficace annuelle que peuvent recevoir les populations en raison de l'exposition au radon, utilisant 68 échantillons différents des matériaux de construction traditionnels les plus utilisés au Yémen et au Maroc. Les concentrations de radon ont été évaluées dans 127 points dans le sol de Qaa Al-Hakel au Yémen et 12 échantillons d'engrais utilisés dans l'agriculture dans cette région.

La technique de la boîte fermée a été utilisée dans cette étude en utilisant des détecteurs de traces nucléaires (CR-39).

Les échantillons locaux et importés ont été collectés à partir de divers matériaux de construction disponibles dans le marché, les carrières, les usines et les maisons en construction.

Les résultats obtenus montrent que, la dose efficace annuelle dans tous les échantillons varie de 18,03 à 127,77 μSv y^{-1}avec une valeur moyenne de 55,74 μSv y^{-1}pour les matériaux de construction au Yémen et de 3,82 à 155,83 μSv y^{-1}avec une moyenne valeur de 27,35 μSv y^{-1} pour les matériaux de construction au Maroc. La dose efficace annuelle pour chaque échantillon a également été déterminée et comparée aux valeurs limites de dose efficace recommandées par le Conseil national de radioprotection et de mesure de 1 à 10 mSv y^{-1}. Les doses efficaces annuelles des matériaux de construction étudiés sont en général faibles et inférieures à la valeur globale.

Les résultats montrent par ailleurs, que le sol de Qaa Al-Hakel contient des concentrations de radon élevées et que les engrais utilisés dans l'agriculture, type de NPK, contiennent de fortes concentrations de radon. L'utilisation de ces engrais entraîne l'accumulation de radionucléides dans le sol, ce qui peut entraîner des risques pour la santé.

La teneur efficace en radium a également été calculée. Toutes les valeurs de la teneur efficace en radium dans tous les échantillons testés se sont révélées être bien inférieures à la valeur admissible.

Mots-clés: Dose efficace, exhalation du radon; CR-39; matériaux de construction

Résumé détaillé

Ce travail de recherche a pour objectif d'évaluer la dose efficace annuelle que peuvent recevoir les populations en raison de l'exposition au radon, utilisant 68 échantillons différents des matériaux de construction traditionnels les plus utilisés au Yémen et au Maroc. Les concentrations de radon ont été évaluées dans 127 points dans le sol de Qaa Al-Hakel au Yémen et 12 échantillons d'engrais utilisés dans l'agriculture dans cette région.

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Les résultats montrent par ailleurs, que le sol de Qaa Al-Hakel contient des concentrations de radon élevées et que les engrais utilisés dans l'agriculture, type de NPK, contiennent de fortes concentrations de radon. L'utilisation de ces engrais entraîne l'accumulation de radionucléides dans le sol, ce qui peut entraîner des risques pour la santé.

La teneur efficace en radium a également été calculée. Toutes les valeurs de la teneur efficace en radium dans tous les échantillons testés se sont révélées être bien inférieures à la valeur proposée par les recommandations internationales.

Cette thèse se compose de quatre chapitres:

Le chapitre 1: décrit les concepts de base relatifs au rayonnement, ainsi que les quantités et unités utilisées en radioprotection en détail.

Le chapitre 2 décrit les concepts relatifs aux propriétés physiques et chimiques du radon, les quantités de base et les terminologies fréquemment utilisées dans les études sur les émissions de radon, le taux d'expiration, le coefficient d'émanation, la diffusion, les isotopes du radon et

sa descendance. De plus, le chapitre 2 présente diverses sources de ce gaz et les mécanismes d'émission, les applications des études sur le radon en géologie et géophysique.

La radioprotection (physique médicale) et les risques radiologiques pour l'homme attribuables à l'inhalation de produits de désintégration du radon ont été discutés en détail, ainsi que les détails des techniques de mesure du radon instantanées et continues, son fonctionnement, ses avantages et ses applications. De plus, au chapitre 2, nous avons discuté des quantités et unités spéciales pour le radon.

Le chapitre 3 décrit les étapes du travail expérimental: collecte et préparation des échantillons, préparation des dosimètres, processus de gravure chimique, puis utilisation de certaines équations pour calculer la dose efficace annuelle, la concentration de radon, le taux d'expiration, la teneur en radium efficace.

Le chapitre 4 décrit les conclusions de l'ensemble des travaux de recherche présentés dans cette thèse de doctorat.

Pour estimer la contribution des matériaux de construction aux concentrations de radon à l'intérieur, une pièce avec un taux d'échange d'air de 0,5 h^{-1} a été considérée. La contribution de la concentration de radon au radon intérieur dans la pièce en raison de l'expiration de la dalle solide d'échantillons de matériaux de construction variait de 0,15 ± 0,03 à 6,18 ± 0,11 Bq m^{-3}, avec une moyenne de 0,93 ± 0,27 Bq m^{-3}. La plus petite concentration de radon accumulée par jour dans une pièce dont le sol et les murs sont entièrement décorés de Ce1, désignant la céramique marocaine, serait de 3,64 Bq m^{-3}, contribuant ainsi faiblement, en moyenne à la concentration de radon dans les maisons marocaines.

En revanche, la plus grande concentration de radon accumulée par jour était de 148,41 Bq m^{-3}pour les échantillons de granite italien. L'échantillon de gravier (Gr2) a montré la valeur la plus basse et la concentration quotidienne de radon accumulée dans une pièce a été estimée à 9,51 Bq m^{-3}.

Pour l'échantillon poudreux et l'échantillon sur plaque, le ciment (C5) et le granit (Gra1) ont tous un niveau d'expiration de radon élevé. Sans ventilation, des niveaux de radon élevés pourraient survenir. En général, les résultats du présent travail se situent dans l'intervalle des valeurs trouvées dans les matériaux de construction, dans les limites de sécurité proposées dans le rapport publié par le Comité scientifique des Nations unies sur les effets des rayonnements atomiques (UNSCEAR 2000).

Il est recommandé que le taux d'expiration du radon soit mesuré pour tous les matériaux de construction et un code standard doit être placé sur tous les produits. Cela minimisera la concentration de radon à l'intérieur dans les nouvelles constructions

Cependant, il existe seulement un petit nombre de maisons qui sont faites de terre seule. Sans ventilation, des niveaux de radon élevés pourraient survenir. En général, tous les résultats pour l'expiration du radon sont inférieurs à la valeur moyenne mondiale de 57.600 Bq m^{-2} h^{-1}.

Pour les matériaux de construction au Maroc, les taux d'expiration de radon de surface à partir des échantillons de dalles solides allaient de 37,92 ± 1,87 mB qm^{-2} h^{-1} à 1545,60 ± 21,26 mB qm^{-2} h^{-1}, avec une moyenne de 232,68 ± 4,25 mB qm^{-2} h^{-1}, où la valeur la plus élevée pour le granit (Gra1), est à peu près égale à cette valeur obtenue dans une étude précédente alors que les taux d'expiration de surface des échantillons poudreux allaient de 99,08 ± 5,06 à 1400,64 ± 31,26 mB qm^{-2} h^{-1}, avec une moyenne de 317,40 ± 4,41 mB qm^{-2} h^{-1}.

Nous avons constaté que la dose efficace annuelle la plus élevée dans les échantillons poudreux du sol yéménite est (91,71 μSv y^{-1}) alors qu'elle est dans le sol marocain (23,645 μSv y^{-1}) (voir Fig. 4-1). Cela est dû au fait que l'échantillon de sol yéménite a été prélevé dans une zone agricole qui est exposée à plusieurs reprises à différents engrais pouvant contenir des quantités d'éléments radioactifs. Les sols étudiés au Maroc appartiennent à la ville de Rabat et prélevés à une profondeur de 30 à 40 cm et, à cette distance le sol aurait pu être exposé au transport d'un endroit à un autre au moment de la construction et donc aux échantillons prélevés sur ces zones ne représentent pas le sol de la ville de Rabat. De plus, le sol de la ville de Rabat n'est pas agricole, donc les engrais n'y sont pas ajoutés de temps en temps. Cette raison nous incite davantage à étudier la concentration de radon dans le sol du Yémen et les engrais utilisés pour aider les cultures à pousser.

D'après notre étude, la dose efficace administrée par tous les échantillons de matériaux de construction étudiés se situe dans la plage du niveau d'intervention recommandé.

- L'un des types de ciment marocains s'est avéré avoir une dose de rayonnement relativement élevée. Il est donc important pour les bâtiments dont les murs sont principalement constitués de ciment de faire particulièrement attention en assurant une ventilation continue et en peignant les murs avec des matériaux qui empêchent le radon de s'échapper dans la maison pour éviter l'exposition au radon.
- Les échantillons de sol dans la zone d'étude du Yémen ont une valeur élevée de dose de rayonnement et cela constitue une menace sérieuse pour la population à l'avenir, en particulier lorsque des engrais sont ajoutés périodiquement.
- Dans les deux études, le granit a une valeur élevée de dose de rayonnement. Il est donc important de limiter ou de réduire l'utilisation du granit, notamment dans les espaces clos. Afin de protéger la population, nous recommandons que le Yémen et le Maroc rejoignent les

pays qui imposent des normes et des lignes directrices sur l'atténuation ou la prévention des risques liés au radon ou fixent des normes d'exposition au radon.

Les résultats de l'étude des niveaux de radon avec la profondeur du sol indiquent que la concentration de radon augmente avec la profondeur pour atteindre la concentration maximale. Cela peut être dû à l'augmentation de la pression et de la teneur en humidité avec la profondeur.

Ainsi, lors de la construction dans des lieux d'étude, il convient de prêter attention aux rez-de-chaussée afin que la réparation des fissures existantes dans le mur et le sol soit reliée directement au sol et donc plus sujette aux fuites et à la collecte du radon. Les rez-de-chaussée doivent être équipés d'un système de ventilation spécial qui élimine constamment le radon.

Concernant le fertilisants, les résultats montrent que les échantillons d'engrais organiques contiennent des concentrations élevées de radon quoique proches de la valeur déterminée au niveau international par la Commission internationale de protection radiologique (CIPR 2014).

Le résultat de notre enquête montre que la plupart des échantillons d'engrais NPK et des échantillons d'engrais complexes contiennent des concentrations de radon élevées qui sont supérieures à la valeur déterminée au niveau international de l'CIPR 2014. L'utilisation de ces engrais doit, si nécessaire, se faire sous stricte supervision et inspection par le ministère de l'Agriculture et le ministère de la Santé, afin d'éviter tout risque sanitaire pour les agriculteurs, les fermes, le sol et les animaux.

Publications and Communication

Part of the results of this work was presented in the form of oral and poster in various national and international conferences, and has been inlcluded in several scientific publications.

Publications:

1) Al Mugahed M, Bentayeb F (2018) Radon exhalation from building materials used in Yemen. Radiat Prot Dosimetry 182:405–412.

2) Al Mugahed M, Bentayeb F (2019) Measurement of radon exhalation rate in various building materials used in Morocco. Int J Low Radiat 11:158–171.

3) Al Mugahed M, Bentayeb F (2019) Studying of radon gas concentrations in soil Qaa Al-Hakel agricultural area, Ibb, Yemen. Mater Today Proc 13:525–529.

Communications:

1) Al Mugahed M, Bentayeb F, The Second International Conference on Materials and Environmental Science (ICMES2018), 26-28 April 2018, Saidia, Morocco.

2) Al Mugahed M, Bentayeb F, The 6^{th} International symposium "Environment and Sustainable Development, 2-4 October, 2019, Faculty of Sciences, Rabt-Agdal, Morocco.

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General Introduction

Radionuclides in building materials are sources of exposure to both external and internal radiation indoors. Exposure to external gamma radiation is caused by the natural radionuclides of building materials, ^{238}U and ^{232}Th and their decay products and ^{40}K. Exposure to internal radiation is caused by the short-lived decay products of radon exhaled from building materials into the indoor air. Materials that contain artificial fallout radionuclides from nuclear weapon tests in the atmosphere or from accidents at nuclear reactors may also exist in building materials.

Radon is a natural radioactive gas resulting from uranium decay. It is an unstable radionuclide that disintegrates through short-lived decay products before eventually reaching the end product of stable lead. The presence of high levels of radon in the indoor and closed environments constitutes a major health hazard to the general population.

The radon progeny is a well-established causative agent of lung cancer and other types of cancers. The lung cancer risk is much higher when radon exposure is combined with smoking. According to the Biological Effects of Ionizing Radiations (BEIR) report, smokers were 10 times more likely to get lung cancer risk than non-smokers.

Recognizing the importance of radon as public health issue, large-scale national and international radon-programs were initiated worldwide. One of such programs is the International Atomic Energy Agency (IAEA) that coordinates the research program 'Radon in the Human Environment', or CRP, involving over 50 countries. Another program is known as the International Radon Project (IRP) by the World Health Organization (WHO) which deals with public health aspects of radon exposure.

IAEA has published a safety guide on Protection of the Public against Exposure Indoors from radon and other natural sources of radiation to assist national authorities in reducing exposure to them. This safety guide also includes guidance on how to prepare a radon action plan.

This research work aims at evaluating the annual effective dose expected to be received by populations due to exposure to radon from 68 different samples of traditional building materials widely used in Yemen and Morocco. It also seeks to evaluate radon concentrations in 127 points in Qaa Al-Hakel soil in Yemen and 12 samples from fertilizers used in agriculture in the same area.

The closed-can technique has been employed in this study using solid-state nuclear track detectors (CR-39). The technique is simple to use and relatively inexpensive and it is the most

widely used method for long monitoring periods because of its good sensitivity, stability against environmental factors and the high degree of optical clarity.

The local and imported samples were gathered from various building materials widely used in Yemen and Morocco. These samples were collected from shops, quarries, factories and homes under construction.

Results obtained from the current study show that the annual effective dose in all samples varies from 18.03 to 127.77 μSvy^{-1}, with an average value of 55.74 μSv y^{-1} for building materials in Yemen and from 3.82 to 155.83 μSv y^{-1}, with an average value of 27.35 μSv y^{-1} for building materials in Morocco.

The annual effective dose for each sample was also determined and compared with the effective dose limit values recommended by the National Council on Radiation Protection and Measurements (from 1 to 10 mSv y^{-1}).

In general, the annual effective doses from the examined building materials are low and under the global value, however, some samples have an average that has relatively high values as in the Yemeni concrete block (CB2), the Yemeni soil (SO2), the Moroccan cement (C5) and some granites in both countries.

The soil of Qaa Al-Hakel agricultural area was chosen to study the radon concentration in detail together with studying the radon gas concentration in the fertilizers used in growing crops. This area is the source of the (SO2) sample which has a relatively high dose.

The results show that the soil contains high radon concentrations which may pose a dangers to the population as this radon moves to buildings through cracks. The results also show that the fertilizers used in agriculture, the NPK fertilizers sample and the complex fertilizer sample, contain high concentrations of radon gas, and this is due to the radionuclides contained in these fertilizers. Use of these fertilizers leads to the accumulation of radionuclides in soil, causing pollution to soil, water drainage systems that can be transmitted to animals and eventually to people who consume the meat or milk from those animals, and hence can lead to health risks.

as Additionally, the effective radium content was calculated. All the values of effective radium content in all samples under test were found to be quite lower than the permissible value of 370 Bq kg^{-1} recommended by the Organization for Economic Cooperation and Development (OECD).

This thesis is presented in four chapters. The highlights of these chapters are given below:

Chapter I: describes the basic concepts relating to the radiation, types of radiation, radioactive decay law, sources of radiation, types of exposure, etc. It also describes the quantities and units used in radiation protection in detail.

Chapter 2 introduces the concepts relating to the physical and chemical properties of radon. The basic quantities and terminologies occurring frequently in radon emission studies, exhalation rate, emanation coefficient, diffusion, radon isotopes and its progeny. It also presents the various sources of this gas and the mechanisms of emission, applications of radon studies in geology and geophysics.

Furthermore, the radiation protection (medical physics) and radon radiological risks to humans attributable to the inhalation of radon decay products were discussed, together with details of radon measurement techniques, instantaneous and continuous, its functioning, advantages, and applications. The special quantities and units for radon were also explained.

Chapter 3 reports on the experimental work stages: collecting and preparing the samples, preparation of dosimeters, chemical etching process, the use of some equations for calculating the annual effective dose, the radon concentration, the exhalation rate, and the effective radium content.

Chapter 4 presents the conclusions of the entire research work undertaken in this PhD. thesis.

The radon contribution to indoor radon, surface exhalation rate, and annual effective doses for the solid slab and powdery samples have been calculated. A comparison of results between Yemen and Morocco was attempted. The radon concentration was also calculated in a horizontal study (50 cm) and concentration with various depths in the soil of the study area as well as the radon concentration in the various fertilizers that are used in that area.

The results obtained in each case have been separately discussed.

It is expected that the experimental data reported here will be useful for radiation protection purposes in order to ensure the radiation safety measures against the possible hazards of inhaling radon and its progeny to people living in the study area. This data will also be a supplement to the radon mapping projects.

CHAPTER 1: RADIATION

1.1 Introduction

Radiation's carriers are electromagnetic waves and/or energetic particles. Basically, radiation can be categorized into two types [1] :

- Non-ionizing Radiation:

Non-ionizing radiation is the term given to radiation in the part of the electromagnetic spectrum where there is insufficient energy to cause ionization. It includes electric and magnetic fields, radio waves, microwaves, infrared, ultraviolet, and visible radiation [2].

- Ionizing Radiation:

Ionizing radiation has so much energy. It can knock electrons out of atoms, a process known as ionization. Ionizing radiation can affect the atoms in living things, so it poses a health risk by damaging tissue and DNA in genes. Ionizing radiation comes from x-ray machines, cosmic particles from outer space and radioactive elements. Radioactive elements emit ionizing radiation as their atoms undergo radioactive decay [3].

1.2 Types of Radiation

There are generally four types of radiation associated with radioactive decay:

1.2 .1 Alpha Particles

An α -particle is a positively charged particle emitted from the nucleus of an atom. It has a mass and charge equal to those of a helium nucleus (two protons and two neutrons) [4].The α-decay occurs mostly in heavy nuclides such as uranium, radon, plutonium, and so forth. Beryllium-8 is the only lightest nuclide that decays by breaking up into two α-particles. After an α-decay, the atomic number of the nucleus is reduced by 2 and the mass number by 4 [5]. The spontaneous emission of an α particle can be represented by the following [6][7]**:**

$$ {}^{A}_{Z}X_{N} \rightarrow {}^{A-4}_{Z-2}Y_{N-2} + \alpha \qquad (1.1) $$

Where *X* and *Y* are the initial and final nuclear species.

All alpha particles emitted by a given radioisotope have the same energy. However, most of the alpha particles that are likely to be found have energies in the range of about 4 to 8 MeV, depending on the isotope from which they came. The alpha particle has an electrical charge of +2. Because of this double positive charge and their size, alpha particles have great ionizing power and, thus, lose their kinetic energy quickly [8]. It is well-known that α-

particles can be stopped by a piece of paper, a few centimeters of air, and gloves. The range of the particles is very short in matter and is approximately 0.03mm in body tissue [5].

These properties cause alpha emitters to be hazardous only if there is internal contamination (i.e., if the radionuclide is inside the body) [8].

1.2.2 Beta Particles

The beta particle has the same mass and charge as an electron. It differs from the electron in its origin. The beta particle, which may be either negatively charged (negatrons) or positively charged (positrons), originates from the nucleus of an atom. The beta particle, like the electron, has a very small mass compared to the proton or neutron. Its mass is 1/1836 that of the proton mass or 1/1838 that of the neutron mass [9].

β Decay: These processes can occur in three possible ways [2]:

- Negatrons (β^-) Decay

When a radionuclide is neutron rich, that is, the N/Z ratio is greater than that of the nearest stable nuclide, it decays by the emission of a β^- particle (note that it is an electron) and an antineutrino. In the decay process, a neutron is converted to a proton, thus raising the atomic number Z of the product by 1. Thus [10][5],

$$n \rightarrow \mathrm{p} + \beta^- + \bar{\upsilon} \qquad (1.2)$$

- Positron (β^+) Decay

When a radionuclide is proton rich, that is, the N/Z ratio is low relative to that of the nearest stable nuclide, it can decay by positron (β^+) emission ccompanied by the emission of a neutrino (υ).

In β^+-decay, essentially a proton is converted to a neutron plus a positron, thus, decreasing the atomic number Z of the daughter nuclide by 1. Thus [5],

$$P \rightarrow n + \beta^+ + \upsilon \qquad (1.3\ )$$

- Electron Capture

A proton-rich radioactive nucleus captures an inner shell orbital electron (usually K shell), transforms a proton into a neutron, and ejects a neutrino. Thus,

$$P + \bar{e} \rightarrow n + \upsilon \qquad (1.4)$$

In this process, the atomic number of the daughter nuclide is lowered by1 [10].

1.2 .3 Electromagnetic Radiations:

Light, radio waves, X-rays and gamma rays are all "electromagnetic" radiation (so called because they have both electric and magnetic properties). In fact, their frequency is the main factor that makes a different from one to another radiation. This is illustrated in Fig. 1-1. X and gamma rays have the highest energies. They cover about the same range of frequencies and, in fact, the only difference between them is their origin [11].

Gamma rays are monochromatic electromagnetic radiations that are emitted from the nuclei of excited atoms following radioactive transformations; they provide a mechanism for ridding excited nuclei of their excitation energy without affecting either the atomic number or the atomic mass number of the atom [12], while X-rays are emitted with discrete energies by electrons as they shift orbits following certain types of nuclear decay processes. A continuous energy spectrum of x-rays called bremsstrahlung may also be emitted by charged particles(e.g. Beta particles) as they decelerate near atomic nuclei [13].

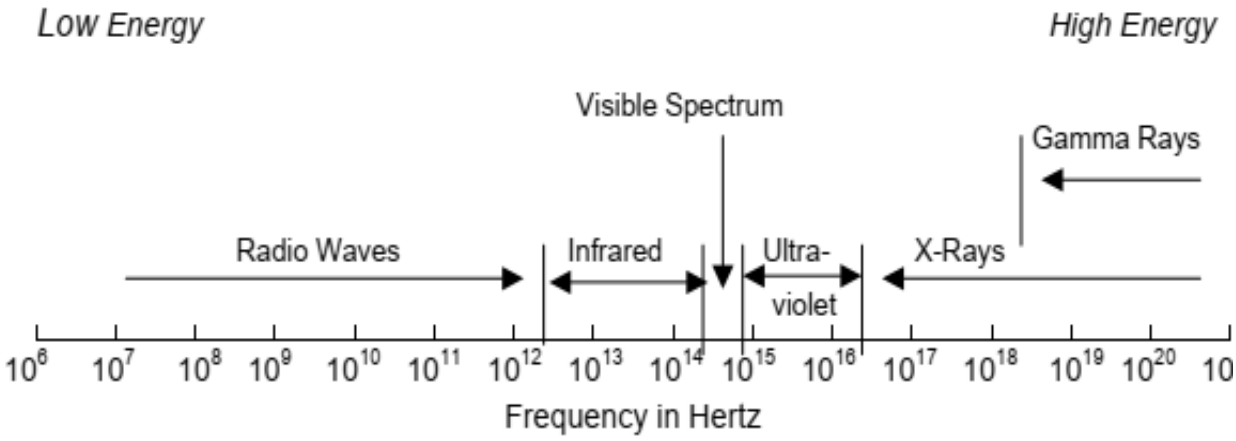

Fig. 1- 1:Electromagnetic Spectrum[11].

1.2.4 Neutrons:

A neutron has no charge and has a mass that is equal to a proton mass. One of its primary purposes is to act as “glue” that holds the nucleus together. If neutrons were not present in the nucleus, the repulsive force between the positively charged particles would cause the nucleus to come apart [14].

Neutrons are typically produced by one of three methods. Large amounts of neutrons

are produced in nuclear reactors due to the nuclear fission process. High energy neutrons are produced by accelerating deuterons and causing them to interact with tritium nuclei. The third method of producing neutrons is by bombarding beryllium with alpha particles [13].

1.3 Radioactive Decay Law

A macroscopic sample of any radioactive isotope consists of a vast number of radioactive nuclei. These nuclei do not all decay at one time, assuming that each nucleus has the same probability of decaying in each second that it exists.

The number of decays that occur in a very short time interval is then proportional to the total number N of radioactive (parent) nuclei present [15].

Consider a system of particles, N_0 in number at time, t = 0 and assume that N is large enough, that we may use calculus. Since particles are integral quantities. Thus, the change in N given by [16]:

$$dN = -\lambda N dt \quad ; N(0) = N_0$$

$$\frac{dN}{N} = -\lambda dt \qquad (1.5)$$

In these equations, λ is a measurable constant called the decay constant, which is different for different isotopes. The greater is, the greater the rate of decay, and the more "radioactive" that isotope is said to be

$$d[log\ N] = -\lambda dt$$

$$log\ N - log\ N_0 = -\lambda t$$

$$log\ N = log\ N_0 - \lambda t$$

$$N = N_0 e^{-\lambda t} \qquad (1.6)$$

Thus, we have derived the well-known *exponential decay law*, $N(t) = N_0 e^{-\lambda t}$.

1.3.1 Half-Life

We can derive the Half-Life Formula starting from Eq. 1.6 by setting [15]:

$$\mathrm{N} = \frac{N_0}{2} \text{ at } \mathrm{t} = T_{1/2}$$

$$\frac{N_0}{2} = N_0 e^{-\lambda T_{1/2}}.$$

So

$$\frac{1}{2} = e^{-\lambda T_{1/2}}$$

$$2 = e^{\lambda T_{1/2}}$$

We take natural logs of both sides ("ln" and "*e*" are inverse operations, meaning ln (e^{x}) and find

$$ln\,(e^{\lambda T_{1/2}}) = ln2,$$

so

$$\lambda T_{1/2} = ln\,2 = 0.693$$

and

$$T_{1/2} = ln\,\frac{2}{\lambda} = \frac{0.693}{\lambda} \qquad (1.7)$$

1.3.2 Mean life Time

Another useful time scale for describing decays is the average or *mean life time* of a radioactive material (τ) which is defined as the average time that a nucleus is likely to survive before it decays, and it can be shown that it relates to the decay constant (λ) in the following way[17]:

$$\tau = \frac{1}{\lambda} \qquad (1.8)$$

1.4 Activity

The activity of a radioisotope is directly related to the number of atoms, or Activity = λN and since N is determined from the mass m of the sample by Avogadro's number, mass can be used directly as a surrogate for activity [18]:

$$m(t) = m_0 e^{-\lambda t} \qquad (1.9)$$

1.5 Sources of Radiation

Environmental radiation originates from several naturally occurring and man-made sources.

1.5.1 Natural Radiation Sources

Most of the radiation that every living being receives comes from natural sources. Man has coexisted with nature from the beginning of his existence, and consequently, has also interacted with radiation and evolved in its presence. The sources of natural radiation can be divided into three types:

1.5.1.1 Primordial Radionuclides

Primordial radionuclides originate mainly from fusion reactions in the interior of supernovae. They have been present since the formation of the planet 4.6×10^9 years ago. Because their half-lives are so long (comparable to the age of the earth), most of them have not yet completely decayed and can be detected [19].

Primordial radionuclides include ^{232}Th, ^{40}K, ^{238}U and ^{235}U. The primordial radionuclides are present in bedrock, soil, building materials, water, air and in the human body. The contents of the natural radioactive substances vary widely between different rocks and soil types, due to the different ways in which they were formed [20].

1.5.1.2 Cosmic Radiation

Cosmic rays are very high-energy particles from extraterrestrial sources that bombard the earth. One source is the sun, which mainly emits alpha particles and protons [12].

The other radiation originates beyond our solar system and is called galactic radiation, consisting mainly of a nucleonic component, which in aggregate accounts for 98% of the total, and electrons, which account for the remaining 2%. The nucleonic component is

primarily protons (85.5% of the flux) and alpha particles (~12%), with the remainder being heavier nuclei (~1%) up to that of uranium [21].

The average annual dose from cosmic radiation is about 26 millirems (mrem) per year. For each 100-meter increase in altitude, this annual dose increases by approximately 1.5 mrem. This increase occurs because, as elevation increases, there is less atmosphere to shield the secondary cosmic radiation [22]. Cosmic-ray intensity increases with increasing latitude north and south of the equator because the earth's magnetic field deflects the high-velocity charged particles that are cutting across the magnetic force field [12].

1.5.1.3 Cosmogenic Radiation

Radionuclides formed by the interaction of cosmic rays with atoms in the atmosphere are known as cosmogenic radionuclides [20].

Several radionuclides of cosmogenic origin are produced when high-energy protons (87% of cosmic radiation) interact with constituents of the atmosphere.

Showers of secondary particles, principally neutrons, from such interactions yield a number of such radionuclides, in particular ^{3}H, ^{7}Be, ^{14}C, and ^{22}Na [16].

1.5.2 Man-Made Radiation Sources

At present medical irradiation ranks first in amount among the man-made (artificial) sources of human exposure. Radiation is used in medicine for diagnostic purposes (e.g., x-ray or nuclear medicine examinations) and for the treatment of diseases, mainly cancers. The doses received by patients are extremely variable: from very small, as in many diagnostic examinations, to very high, such as those delivered in clinical radiotherapy [23].

In addition, members of the public are exposed to radiation from consumer products, such as building materials, tobacco (^{210}Po), ophthalmic glass, televisions, luminous watches and dials (tritium) combustible fuels (gas, coal, etc.), airport X-ray systems, smoke detectors (americium), etc. [21][9].

Fig. 1-2 shows the sources of radioactive risk, the relative contributions sources: natural and man–made. It could be seen that natural sources provide about 85%, while the non–natural sources provide 15% of the average total dose that a human being commonly receives [24].

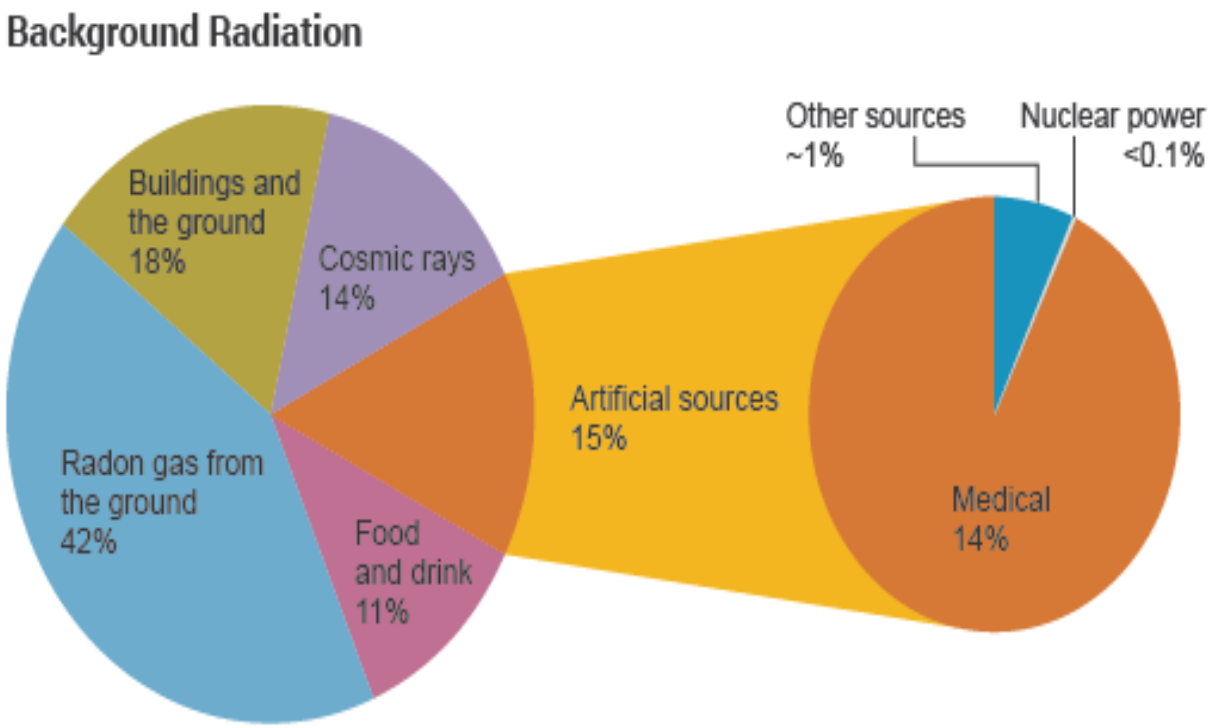

Fig. 1-2:Sources of radiation exposure [24].

1.6 Exposures

There are two principal routes by which humans are exposed to ionizing radiation, external and internal.

1.6.1 External Exposures

External irradiation from radionuclides naturally present in the environment or released from man-made practices or events is usually an important component of the exposure of human populations. These exposures derive primarily from gamma radiation arising from the decay of these radionuclides at locations outside the human body. Secondarily, exposures to the skin from beta radiation may be considered [25].

Beta and alpha radiation from external sources are far less penetrating and deposit their energy primarily on the skin's outer layer. Consequently, their contribution to the absorbed dose of the total body dose, compared to that deposited by gamma rays, may be negligible [8].

As with all radiation exposures, the size of the dose resulting from an external exposure is a function of:

1- Time

The total radiation exposure to an individual is directly proportional to the time of exposure to the radiation source.

2- Distance

The intensity of a radiation source, and hence the radiation exposure, varies inversely as the square of the distance from the source to the point of exposure. It is recommended that an individual should keep as far away as practically possible from the radiation source [26].

3- Shielding

Various high atomic number (Z) materials that absorb radiations can be used to provide radiation protection.

4- Activity

It should be obvious that the radiation exposure increases with the intensity of the radioactive source. The greater the source strength, the more the radiation exposure. Therefore, one should not work unnecessarily with large quantities of radioactivity [5].

1.6. 2 Internal Exposures

Internal exposures are those that result from radioactive material which has been taken up by the body as a result of ingestion, inhalation, injection, or absorption through the skin [27].

Such exposures are of concern for all radioactive materials regardless of the type of ionizing radiation emitted. Of particular concern, however, are radioactive materials which emit alpha and beta particles, because all their energy is dissipated locally in short densely ionizing tracks, these radiations cause significant damage to tissue [13].

The majority of elements are stored in particular organs so that the different dose rates of radioactive intake may affect various organ of the body. For example, iodine is stored in the thyroid gland, and plutonium is stored in the lungs or bones [28].

The most vital organs are reproductive organs, kidneys, lungs, liver, etc., depending upon the nature of radionuclides and the route through which they entered the body. Their effects may cause lung cancer, damage to genetic organs, and eye defects, and destroy the circulatory system [27].

The dose committed to a particular organ or tissue depends, in part, upon the time over which these areas are irradiated by the radionuclide. This, in turn, is determined by the

effective half-life [13], Where the effective half-life (T_e) is defined as: Time required for an initial administered dose to be reduced to one half as a result of both physical decay and biological elimination of a radionuclide. It is given by

$$T_e = \frac{T_p \times T_b}{(T_p + T_b)} \qquad (1.10)$$

where T_e is the effective half-life, and T_p and T_b are the physical and biological half-lives, respectively [5]. The biological half-life of a radionuclide is defined as the time required for a compartment of a biological system to eliminate (in the absence of additional input and radioactive decay) half of its radionuclide content [29].

1.7 Quantities and Units Used in Radiation Protection

Radiation protection is the set of rules, standards, procedures, means of prevention and surveillance aimed at preventing or reducing the harmful effects of ionizing radiation produced on persons, including the damage caused to environment in accordance with the standards prescribed by the laws and regulations[30].

The quantities in radiation protection can be classified into three broad categories:

- Physical quantities;
- The protection quantities;
- Operational quantities [30].

1.7.1 Physical Quantities

Physical quantities are quantities that are accessible quantitatively by calculation or by measurement.

1.7.1.1 Radiometric quantities

These are quantities used to describe external radiation fields. We can cite the flow, fluence and fluence rate [30].

A. Fluence(Φ): The particle fluence Φ is the quotient $d\ N$ by dA, where dN is the number of particles incident on a sphere of cross-sectional area dA:

$$\Phi = \frac{dN}{dA} \qquad (1.11)$$

The unit of particle fluence is m^{-2}. The use of a sphere of cross-sectional area *dA* expresses in the simplest manner the fact that one considers an area *dA* to be perpendicular to the direction of each particle and hence that particle fluence is independent of the incident angle of the radiation.

Fluence rate($\dot{\Phi}$): The particle fluence rate $\dot{\Phi}$ is the quotient of $d\Phi$ by dt, where $d\Phi$ is the increment of the fluence in time interval *dt*:

$$\dot{\Phi} = \frac{d\Phi}{dt} \qquad (1.12)$$

with units of $m^{-2}s^{-1}$ [31][32].

1.7.1.2 Dosimetric Quantities

Dosimetric quantities describe the processes by which particle energy is converted and finally deposited in matter [32], which quantify the exposure of humans to ionising radiation appropriately for implementation of the fundamental principles of limitation and optimization [33][34].

1-Absorbed dose (*D*)

The physical dose quantity is given by:

$$D = \frac{d\acute{\varepsilon}}{dm} \qquad (1.13)$$

Where $d\acute{\varepsilon}$ is the mean energy imparted by ionising radiation to matter in a volume element, and *dm* is the mass of matter in this volume element. The SI unit for absorbed dose is joule per kilogram (J/kg) and its special name is gray (Gy) [33], [35], [36].

2-**Kerma** (*K*)

The quotient of dE_{tr} by dm , where dE_{tr} is the mean sum of the initial kinetic energies of all charged particles in a mass dm of a material by the uncharged particles incident on dm, thus:

$$k = \frac{dE_{tr}}{dm} \qquad (1.14)$$

The unit of kerma is $J\ kg^{-1}$, and its special name is gray (Gy) [32], [36], [37].

1.7.2 Protection Quantities

The protection quantities, equivalent dose in a tissue or organ and effective dose, were developed by the International Commission on Radiological Protection (ICRP) to allow quantification of the extent of exposure of the human body to ionising radiation. These quantities are used for the implementation of limitation and optimisation principles[38]. Body-related protection quantities are not directly measurable [39][38].

1.7.2.1 Equivalent Dose (H_T)

To reflect the damage done in biological systems from different types of radiation, the equivalent dose is used [40].

The equivalent dose, $H_{T,R}$, in tissue or organ T due to radiation R, is given by[41][35][42]:

$$H_{T,R} = w_R D_{T,R} \qquad (1.15)$$

Where $D_{T,R}$ is the average absorbed dose from radiation R in tissue T and w_R is the radiation weighting factor.Values of w_R are given in Table 1-1

Table 1- 1:Radiation weighting factors, w_R [33][43] [34].

Radiation type*	Energy range	Radiation weighting factor, w_R
Photons	All energy	1
Electrons and muons	All energy	1
Protons and charged pions	(All energy)	2
Alpha particles, fission fragments, heavy ions	All energy	20
Neutrons	E_n<1 MeV	$2.5 + 18.2\, e^{-(\ln(E_n))^2/6}$
	1 MeV $\leq E_n \leq$ 50 MeV	$5 + 17.0 e^{-(\ln(2E_n))^2/6}$
	E_n > 50 MeV	$2.5 + 3.25 e^{-(\ln(0{,}04E_n))^2/6}$

*All values relate to the radiation incident on the body or, for internal sources, emitted from the source.

Equal equivalent doses from different sources of radiation delivered to a point in the body should produce approximately the same biological effect. However, a given equivalent dose will in general produce different effects in different parts of the body. A dose to the hand is, for example, considerably less serious than the same dose to blood forming organs [40].

If an organ is irradiated by more than one type of radiation, the equivalent dose is given by the sum [31][26][40][44][38]:

$$H_T = \sum_R (w_R D_{T,R}) \qquad (1.16)$$

The SI unit of equivalent dose is J/kg and its name is the sievert (Sv); the old unit is the rem and the relationship between the two units is 1 Sv = 100 rem

1.7.2.2 Equivalent of effective dose or effective dose

Each tissue or organ has its own radio sensitivity. Thus for the same equivalent dose (H_T) we have different risks and biological effects depending on the tissue or the irradiated organ.

This is why the ICRP introduced a magnitude characterizing the effect of radiation on tissues and organs called effective dose *E*.

The effective dose of radiation received by a person is, in simple terms, the sum of the equivalent doses received by all tissues or organs, weighted for "tissue weighting factors." These reflect different sensitivities to radiation of different organs and tissues in the human body. The SI unit for the equivalent and effective dose is the sievert (Sv), where 1 Sv = 1 J/kg [45]. The formula follows the expression of the effective dose [25][46][47][48][49]:

$$E = \sum_T w_T H_T \qquad (1.17)$$

Where H_T is the equivalent dose in tissue or organ T and w_T is the tissue weighting factor. The ICRP tissue weighting factors are shown in Table 1-2.

Table 1-2: ICRP Tissue weighting factors [34][29].

Tissue or organ	**W_T given in %** w_T
Gonads	0.08
Bone marrow (red)	0.12
Colon	0.12
Lung	0.12
Stomach	0.12

Bladder	0.04
Breast	0.12
Liver	0.04
Esophagus	0.04
Thyroid	0.04
Skin	0.01
Bone area	0.01
Brain	0.01
Salivary glands	0.01
Other tissues or organs*	0.12

* Remainder tissues: Adrenals, Extra thoracic (ET) region, Gall bladder, Heart, Kidneys, Lymphatic nodes, Muscle, Oral mucosa, Pancreas, Prostate (♂), Small intestine, Spleen, Thymus, Uterus/cervix (♀).

1.7.2.3 Committed dose

When radionuclides are taken into the body, the resulting dose is received throughout the period of time during which they remain in the body. The total dose delivered during this period of time is referred to as the committed dose.

The committed dose may refer to the committed effective dose and the committed equivalent dose [31][25].

The committed equivalent dose, $H_T(\tau)$, in a tissue or organ T is defined by [46]:

$$H_T(\tau) = \int_{t_0}^{t_0+\tau} H_T(t)dt \qquad (1.18)$$

Where τ is the integration time following the intake at time t_0. The quantity committed effective dose $E(\tau)$ is then given by:

$$E_T(\tau) = \sum_T W_T H_T(\tau) \qquad (1.19)$$

The SI unit of E_T (τ) is the sievert (Sv) or but rem (roentgen equivalent man) is still commonly used (1 Sv = 100 rem).

1.7.3 Operational quantities

Operational quantities are dose quantities defined for use in radiation protection measurements for external exposure. with the objective of "reasonable" estimation of the protection quantities because protection quantities generally are not measurable [50], there are two types of operational quantities intended to:

- The monitoring of work areas and the environment

- Individual monitoring

1.7.3.1 Monitoring of Work Areas and the Environment

For the monitoring of work areas and the environment, there are two quantities, namely, the ambient dose equivalent, $H^*(d)$, and the directional dose equivalent, $H'(d, \Omega)$.

1-Ambient dose equivalent *H*(d)*

The ambient dose equivalent, $H^*(10)$, defined at a point in a radiation field, is the dose equivalent, which would be produced by the corresponding expanded and aligned field in the sphere of International Commission on Radiation Units and Measurements (ICRU) at a depth of 10 mm on the radius opposing the direction of the field [34]. The ICRU sphere is a 30 cm diameter tissue-equivalent sphere, with a composition of 76.2% oxygen, 11.1% carbon, 10.1% hydrogen, and 2.6% nitrogen. A depth d = 10 mm is recommended for strongly penetrating radiation [51][52][53].

2-The directional dose equivalent, *H'(d, Ω),* at a point of interest in the actual radiation field, is the dose equivalent that would be produced by the corresponding expanded radiation field, in the ICRU sphere at a depth d, on a radius in a specified direction Ω [54].The recommended values of d are chosen for the assessment of various doses: d = 10 mm for effective dose, d = 3 mm for dose to the lens of the eye, and d =0.07 mm for dose to the skin and to the hands and feet. The unit of ambient dose equivalent, directional dose equivalent, and personal dose equivalent is J kg^{-1} or Sv[38].

1.7.3.2 Personal Dose Equivalent

The personal dose equivalent, $H_p(d)$, is the operational quantity for individual monitoring: the dose equivalent in soft tissue (ICRU-sphere) below a specified point on the body at an appropriate depth d. This quantity can be used for measurements of superficial and deep organ doses. For superficial organs, depths of 0.07 mm for skin and 3 mm for the lens of the eye are employed, and the personal dose equivalents for those depths are denoted by

$H_p(0.07)$ and $H_p(3)$, respectively. For deep organs and the control of effective dose, a depth of 10 mm is frequently used, with the notation $H_p(10)$ [43].

1.7.3.3 Operational Quantities and Annual Dose Limits

Since operational quantities are good estimators of protection quantities, they can be related to annual effective and equivalent dose limits. Fig.1-3 and Table 1-3 respectively summarize the relationships between different quantities of radiation protection and the regulatory limits for exposure doses.

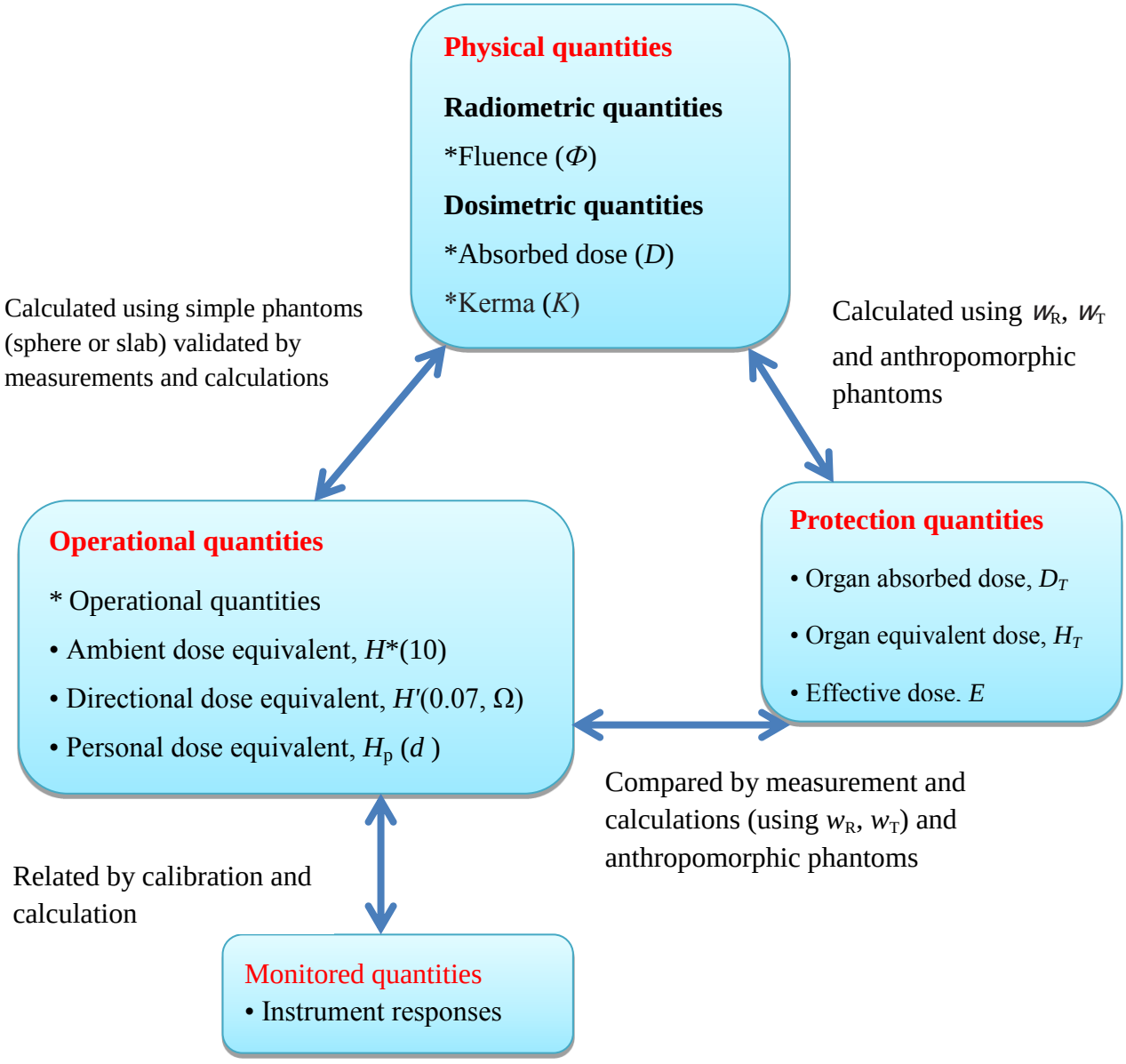

Fig. 1-3: Relationship between physical protection and operational quantities [55].

Table 1-3: The recommended dose limits, stated by the ICRP in ref [34] **except for the eye lens dose which have been revised in ref.**[56].

Occupational	Protection quantities		operational quantities	limit
	Effective dose (D)		$H_p(10)$	20 mSv/year on average 5 years
	Equivalent Dose	Skin, hands, feet	$H_p(0,07)$	500 mSv/year
		Lens of the eye	$H_p(3)$	20 mSv/year
Public	Effective dose (D)		$H_p(10)$	1 mSv/year on average 5 years
	Equivalent Dose	Skin, hands, feet	$H_p(0,07)$	50 mSv/year
		Lens of the eye	$H_p(3)$	15 mSv/year

CHAPTER 2: RADON

2. 1 Radon History

In 1900 Ernst Freidrich Dorn, a German physicist established that radium emitted a radioactive gas which we now call radon [57]–[60].

J. Thomson observed the existence of radon in water in 1902. He was performing a series of experiments on the electrical conductivity produced in gases when passing through water. Subsequently in 1904, H.F.R. von Traubenberg demonstrated that radon exist in the tap water of the Freiburg city (Germany), which was followed by the discovery of radon in groundwater [57], [61].

In 1908, William Ramsay and Robert Whytlaw-Gray isolated radon and reported the value of radon density which was the heaviest known gas [62].

Radon (^{222}Rn) was accepted in 1912 as a new element by the International Commission for Atomic Weights [59], [63].

in 1923, the International Committee for Chemical Elements and the International Union of Pure and Applied Chemistry (IUPAC) approved the use of the names radon (Rn), thoron (Tn), and actinon (An) for the three isotopes of this element with the longest half-lives [59], [63], [64]. For many years, the radon continued in some scientific circles to be called radium emanation or simply "emanation". As early as 1901 the presence of radioactive substances in the air was established by collecting activity on a wire suspended in outdoor air at a high negative potential [59].

2.2 Physical and Chemical Properties of Radon

2.2.1 Chemical Properties of Radon

Radon is one of the noble gases. Radon atoms have a closed-shell electronic structure and are extremely stable, as the ionization enthalpies are high. There are no ordinary electron-pair interactions among the noble gas atoms [65].

Radon lies on a diagonal between the true metals and non-metals in the periodic table. Therefore, it is referred to as a metalloid element. In fact, Radon has some of the characteristics of both groups and behaves similarly to some elements such as boron, germanium, antimony and polonium [66], [67].

Radon can react with several complexes (such as fluorine, halogen fluorides, dioxygenyl salts, fluoronitrogen salts, and halogen fluoride-metal fluoride complexes) to form

ionic compounds. Radon can be collected by using several solid reagents but must be protected from moisture, since they hydrolyze readily. Recently, solutions of nonvolatile, cationic radon have been produced in nonaqueous solvents [66].

In addition, radon can be quantitatively collected on ion exchange columns packed with either nation resins or complex salts. In its ionic state, radon is able to displace H^+, Na^+, Cs^+, Ca^{2+} ions from a number of solid materials [64], [66].

Radon reacts spontaneously at room temperature with many solid compounds that contain oxidizing cations, such as. $Cl\,F_2^+$, $Br\,F_2^+$, $Br\,F_4^+$, O_2^+ , and N_2F^+[68].

Radon is slightly soluble in water and about 16 times soluble in fats and organic liquid [69], with the exception of glycerine that holds a lower solubility than in water [70]. And its solubility depends on water temperature where the greater solubility appears on the lowest temperature of water [69], [71], as shown in Fig. 2-1.

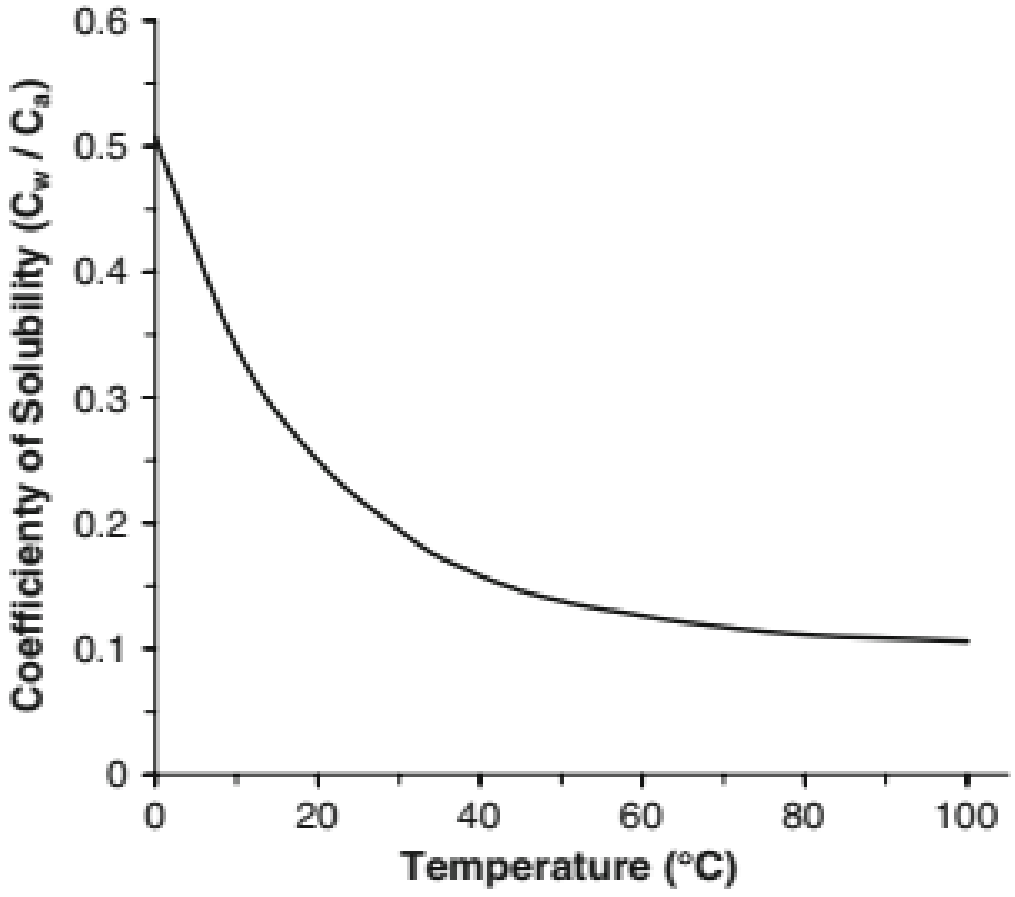

Fig.2-1: Solubility of radon in water with respect to temperature (where C_w and C_a are concentrations in water and air, respectively) adapted from [57].

Radon is readily absorbed on charcoal and silicon gel [63], [70]. In a multiphase system at normal environmental temperature, radon concentrations are greatest, intermediate and least in the organic liquids, gas and the water phases, respectively [70], [71]. If temperature is increased, the concentration in the gas phase increases at the expense of the liquid phase [70]. These properties have been successfully exploited for the determination of radon gas and for its extraction from other gases and/or liquids [71].

2.2.2 Physical Properties of Radon

Radon is a naturally occurring, colorless, odorless, and radioactive gas. Compared to the other noble gases, radon is the heaviest and has the highest melting point, boiling point, critical temperature, and critical pressure [57].

When cooled below its freezing point, radon has brilliant phosphorescence. It becomes yellow at lower temperatures and orange-red at the temperature of liquid air. Because of this property , radon was called niton (the shining one) at the time of its discovery by Dorn in 1900 [64], [67]. The main physical, chemical, and radiological properties of radon are listed in Table 2-1

Table 2-1:Physical, Chemical, and Radiological Properties of Radon [72]**.**

Property	**Values**
Atomic number	86
Standard atomic weight	222
Outer shell electron configuration	6s26p6
Density	9.73 kg m^{-3} (at 0 °C, 1.013×10^5 Pa)
Melting point (°K)	202
Normal boiling point (°K)	208.2
Heat of fusion (kJ mol^{-1})	3.247
Heat of vaporization (kJ mol^{-1})	18.0
First ionization enthalpy (kJ mol^{-1})	1037
Oxidation states	0, 2, 6
Electronegativity	2.2 (Pauling scale)
Covalent radius (nm)	0.150
van der Waals radius (nm)	0.220
Half-life $T_{1/2}$	3.823 d
Decay constant λ	2.098×10^{-6} s^{-1}
Diffusion coefficient in air D_a	1×10^{-5} m^2 s^{-1}
Diffusion coefficient in water D_w	1×10^{-9} m^2 s^{-1}

2.3 Factors Controlling Radon Releases

Releasing of radon from a residue repository to the atmosphere take place by the following series of processes:

(a) Emanation: radon atoms formed from the decay of radium escape from the grains (mainly because of recoil) into the interstitial space between the grains.

(b) Transport: diffusion and advective flow cause the movement of the emanated radon atoms through the residue or soil profile to the ground surface.

(c) Exhalation: radon atoms that have been transported to the ground surface and then exhaled to the atmosphere [73]. These processes are illustrated in Fig. 2-2**.**

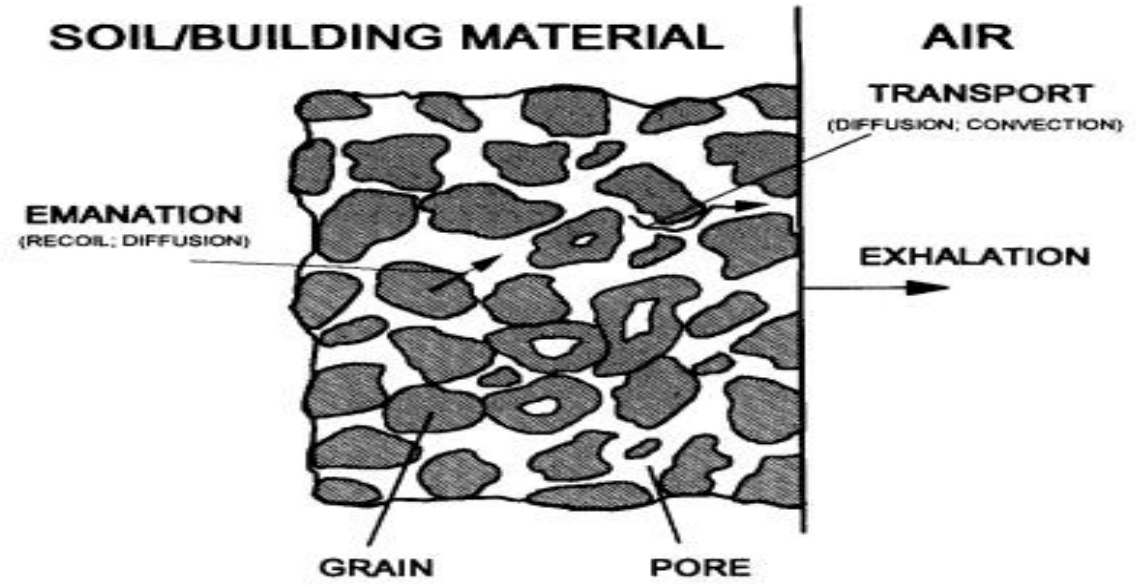

Fig.2-2:Mechanisms of exhalation of radon from soil/ building material [74].

2.3.1. Emanation

The released fraction of radon atoms into a rock or soil pore space from a radium-bearing grain is termed the ‘radon emanation coefficient’, ‘emanating power’, or ‘escape-to production ratio’ of the soil. The radon emanation coefficients of typical rocks and soils range from 0.05 to 0.70 [75], [76]. The emanation fraction has two components: recoil and diffusion. Because of the low diffusion coefficient of gases in the solid grain material, it is assumed that the main fraction of the emanation power is contributed by the recoil process. After the alpha decay of the radium isotopes (^{226}Ra, ^{224}Ra), radon and thoron atoms possess kinetic energies of 86 and 123 keV, respectively [74], [77], [78].

This energy can be calculated on the basis of the law of conservation of linear momentum. The main factors that determine whether the newly formed radon can escape to pore spaces (emanation: points A, B, E and F in Fig. 2-3) or stay in the grain (not emanation: points C, D and G in Fig. 2-3) are the birthplace of radon in a grain, recoil direction, etc. [79].

The distance of the recoil travels depends upon the composition and density of the materials that encounters [80][75]. The range of recoil distance for ^{222}Rn is 20-70 nm in common minerals [78][75], 100 nm in water and 63 μm in air [25].

Only radium atoms within the recoil range from the grain surface can produce radon atoms that have any possibility of being emanated. Even if radon was released from a radium-bearing grain, it can penetrate the fluid-filled pore space, depending on its residual energy, to collide with a neighboring grain. In this case, radon can be embedded with the threshold energy [81].

After the embedding, radon may migrate from the pocket created by its recoil passage into pore (point E in Fig. 2-3); or decays radioactively after the molecular diffusion in the grain (points D and G in Fig. 2-3). The former contributes to the emanation, but the latter not. On the other hand, radon completely escaping into inner pore space in the grain must diffuse to outer pore (point F in Fig. 2-3). For the emanation, the radon atoms that cannot diffuse out into the outer pore or are adsorbed on the inner surface of the grain should be regarded as being not emanated [79].

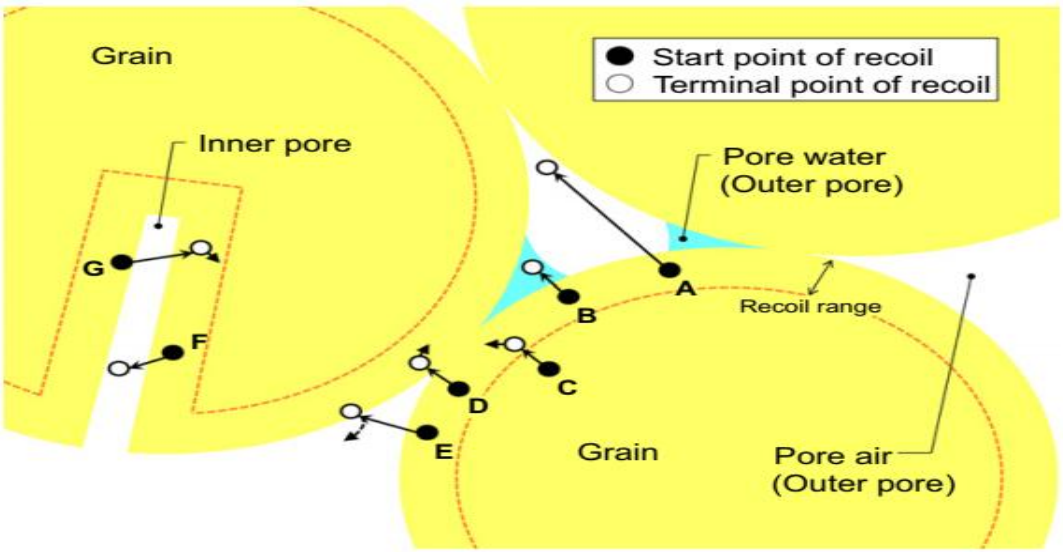

Fig.2-3: Scheme of radon emanation phenomenon. Emanation: (A), (B), (E) and (F). No emanation: (C), (D) and (G). If radon cannot diffuse out from inner pore into outer, radon in point (F) should not be regarded as being emanated. Arrows following terminal points of recoil represent diffusion process, which are not to scale [79].

2.3.1.2 Factors Affecting Radon Emanation

The emanation coefficient of materials was affected by many factors which are related to the solid grain properties and the pore space properties such as radium distribution inside solid grains, moisture content, soil temperature, and solid grain size. The relation between the emanation coefficient and these factors has been described in detail as follows.

2.3.1.2 .1 Radium Distribution, Particle Size and Shape

Determine in part how much uranium and radium is close enough to the surface of the grain to allow the radon to escape into the interstitial pores. If radium were uniformly distributed throughout a grain, the emanation coefficient would approach an inverse proportion to the particle diameter if this were greater than approximately 0.1 μm. On the other hand, if radium were mainly distributed on the grain surface, the emanation coefficient would be constant regardless of the particle diameter (See Fig. 2-4.) [72].

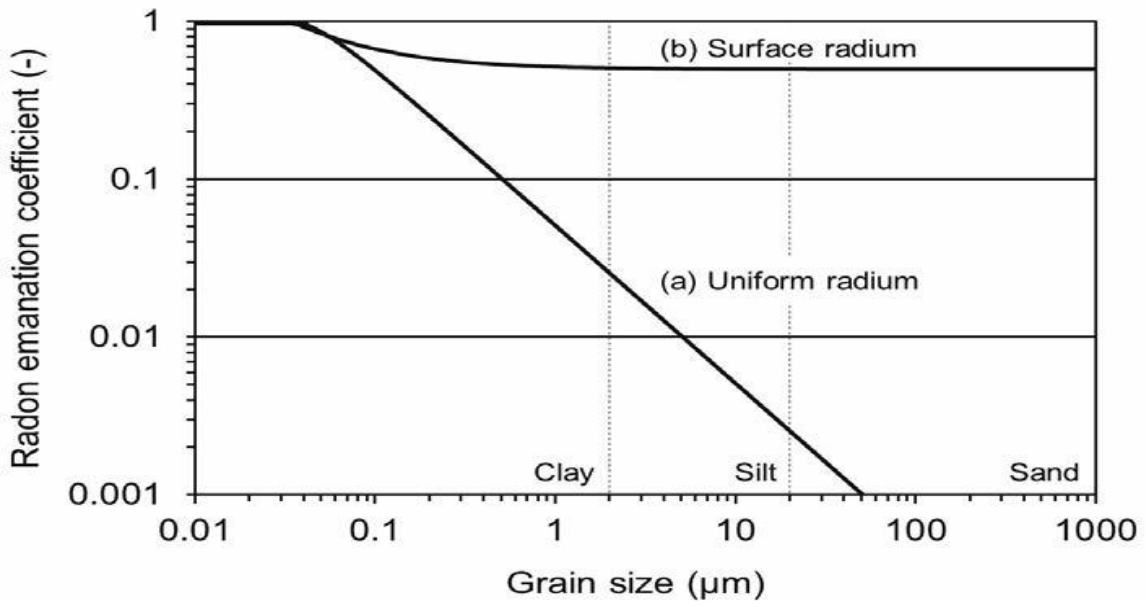

Fig.2-4: Calculations of radon emanation coefficients in the single grain model. Radium is assumed to be distributed (a) uniformly in a spherical grain, and (b) on the surface of a spherical grain. This model does not take into account the embedding of radon into a second grain. The range of radon with a recoil energy of 86 keV in a common mineral (quartz, SiO2) was set to 34 nm [72].

2.3.1.2 .2 Moisture Content

Several studies have shown that moisture saturation is positively correlated with the radon emanation rate [82][83][84][85].The explanation for this phenomenon seems to lie in the lower recoil stopping distance for radon in water than in air. A radon atom entering a pore that is filled or partially filled with water has a higher probability of being stopped in the pore volume without crossing the pore space and penetration another grain [74][85].

High moisture levels decrease the radon emanation rate because of slowed diffusion [86][78][87] as shown in Fig. 2-5 [79]. As moisture content was increased, the radon emanation fraction increased to reach a peak, and then decreased.

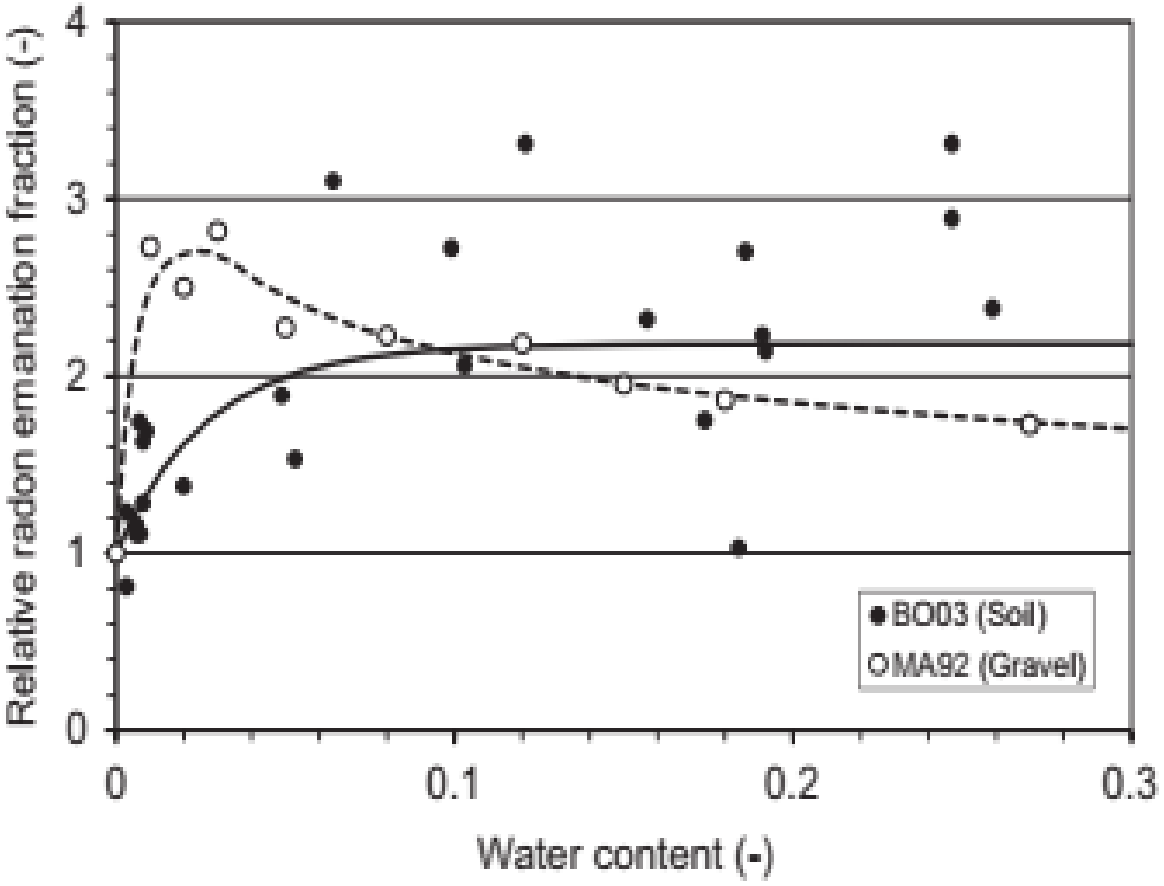

Fig.2-5:Relative radon emanation fraction as a function of moisture content [79].

3.1.2 .3 Temperatures

The reduction in physical adsorption of radon onto grains that occurs during the diffusion, is the reason of increase in the radon emanation coefficient at high temperature [82][88].

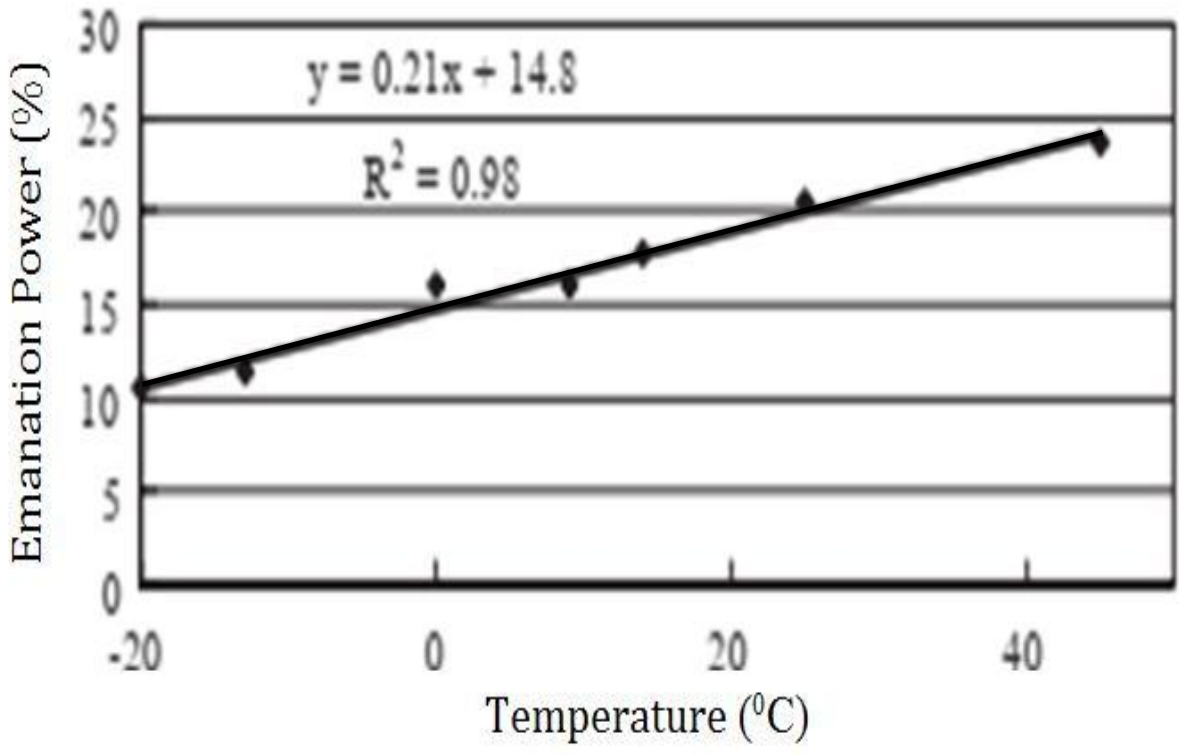

Fig.2-6:The correlation between soil temperatures and radon emanation power [88]**.**

2.3.2 Diffusion

The phenomenon that is called molecular or atoms diffusion describes the migration of the radon gas toward the direction of its decreasing concentration in the air which results from the random movement of radon gas atoms mixed in the air [89], [90]. The migration takes place in the medium, be it solid, liquid, or gas [57].

Diffusion coefficients of radon in solids are extremely low [76][91], and the radon will decay before moving any distance, regardless of crystal size. Transport by molecular diffusion of radon in soils is limited because of its short half-life. Migration by diffusion ranges from about 5 m in gravel to about 2 cm in saturated mud or clay, and distances greater than 1 m are probably unusua1 [57].

2.3.3 Exhalation Rate

The radon exhalation rate is defined as the amount of activity released per unit surface area per unit time from the material. It depends on the ^{226}Ra content of the material, emanation coefficient, gas diffusion coefficient in the material, porosity and density of the material [92][93].

The emanation factor and the diffusion coefficient depends on the physical properties of building materials or soil such as spatial distribution of radium atoms in the mineral grain, texture, humidity content, porosity and density [94] . Also, there are many external factors that can influence the diffusivity and thereby the exhalation rate: rainfall, snowfall, freezing and increasing atmosphere pressure decrease the exhalation rate, while increasing wind speed and temperature can increase it. In addition, these meteorological parameters can cause a convective air flow in the pore space and change the radon flux from the soil [74].

Mechanisms of exhalation of radon from soil/building material are shown in Fig. 2- 2[74].

2.4. Radon Isotopes Diffusion Coefficient

Uranium and thorium are present everywhere in the Earth's crust in varying amounts. These radioactive elements are sources of three radioactive decay series. Radon gas is one of the decay products of these series [93].

There are 35 known radon isotopes (all radioactive) with atomic mass numbers ranging from 195 to 229 [95].

However there are three major isotopes of radon, namely: ^{222}Rn (called radon, belongs to ^{238}U decay series); ^{220}Rn (called thoron, belongs to ^{232}Th decay series) and ^{219}Rn (called actinon, belongs to, ^{235}U decay series) See Fig. 2-7, 2-8 and Fig. 2-9.

^{222}Rn has 3.82 days half-life, whereas^{220}Rn (55.6 s) and ^{219}Rn (3.96 s) have much shorter half-lives than ^{222}Rn. That is why ^{220}Rn and^{219}Rn are given less importance in environmental studies. Scientifically, radon is known to be ^{222}Rn, the most abundant isotope of the element radon [93], [96].

2.5. Radon Progeny

Radon progeny are sometimes referred to as 'radon decay products'[97], [98] or the more historic term 'radon daughters' [99], [100]. ^{222}Rn decay products are divided into two groups, namely, the short-lived (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po) and the long-lived (^{210}Po, ^{210}Pb, and ^{210}Bi) daughters. Since the longest-lived element of the first group has a half-life of less than 27 min (See Fig. 2-7), the whole sequence of decays can be completed before the human clearance processes can sweep them away.

The long-lived ^{222}Rn progeny contributes relatively little to lung exposure because the first nuclide, ^{210}Pb, of this group has a large half-life (22.3 y) so that is utterly removed from the body before decaying [49].

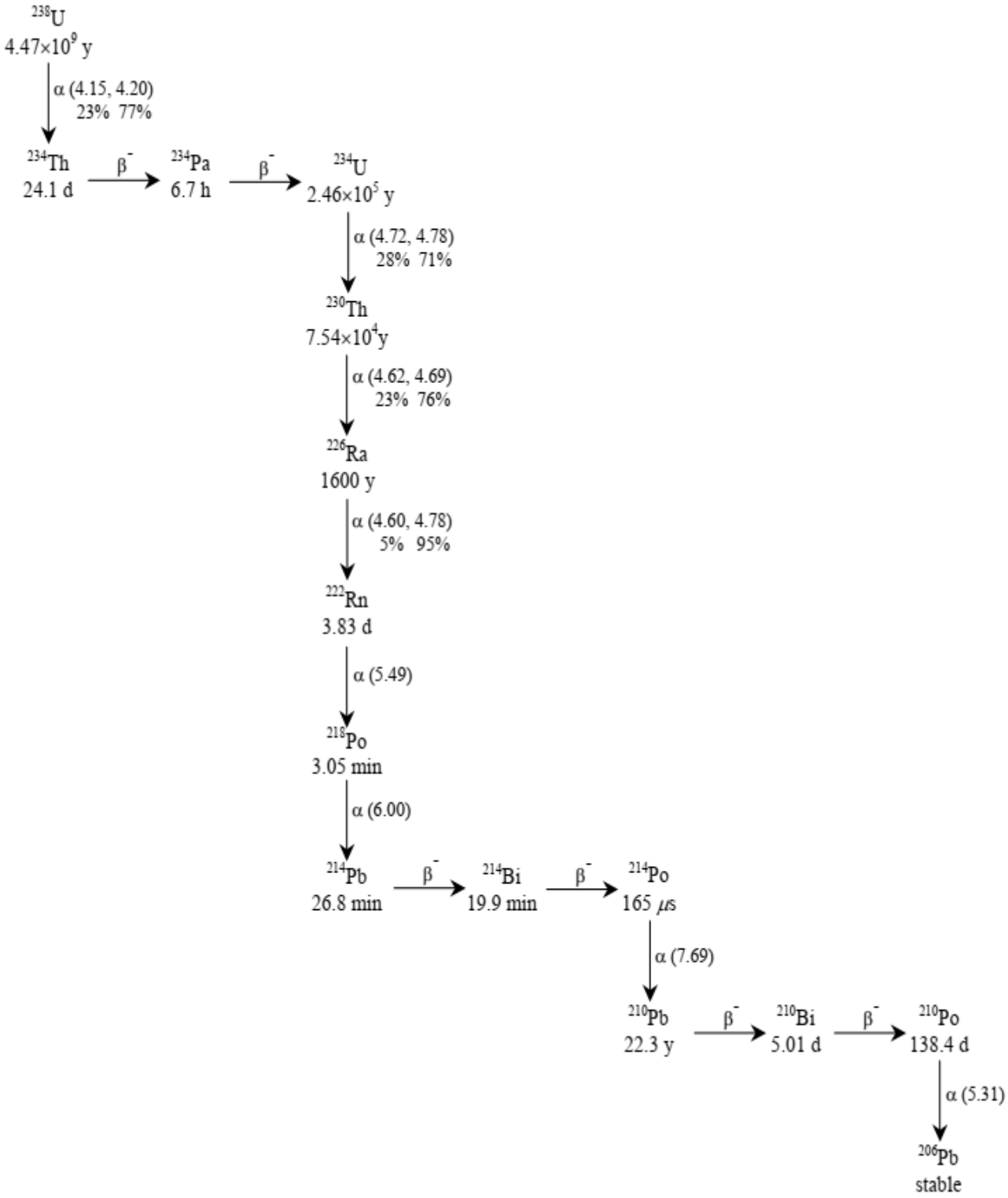

Fig.2-7:Decay diagram of ^{238}U series with the half-life of each radionuclide and the energies of α-emissions expressed in MeV [101]**.**

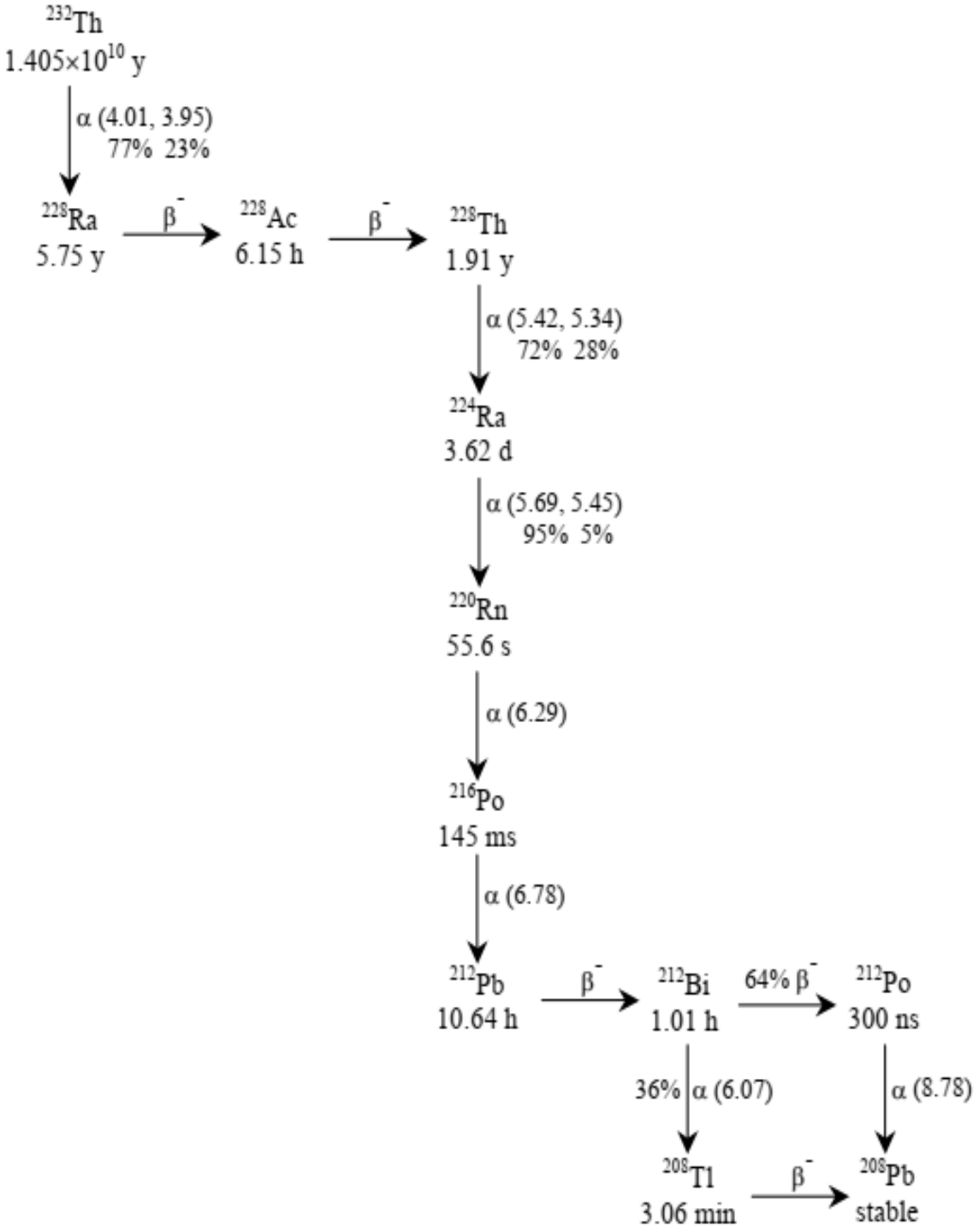

Fig.2-8: Decay diagram of ^{232}Th series with the half-life of each radionuclide and the energies of α-emissions expressed in MeV [101].

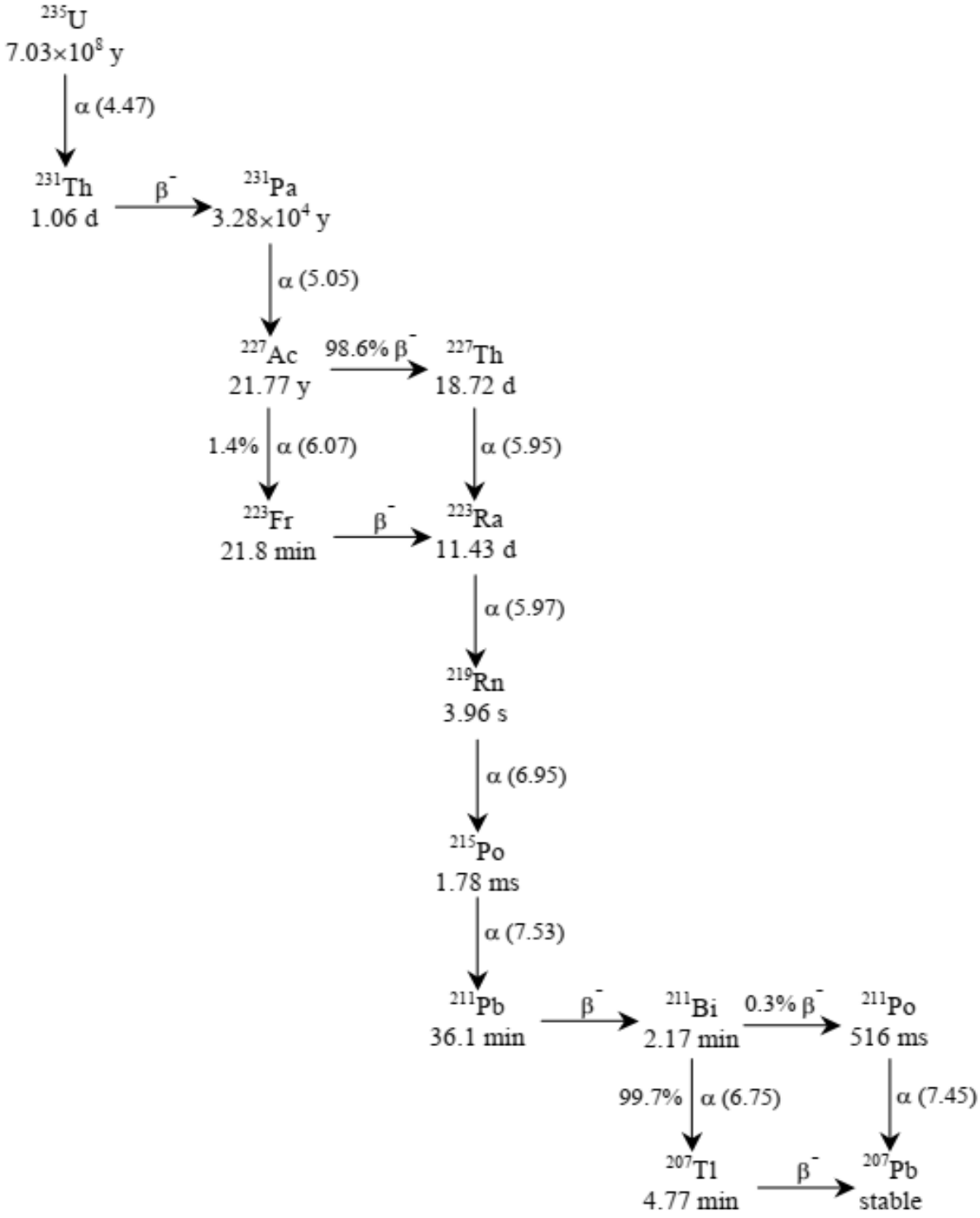

Fig.2-9: Decay diagram of ^{235}U series with the half-life of each radionuclide and the energies of α-emissions expressed in MeV [101]**.**

2.6 Sources of Radon

The sources of radon include the materials containing radium content such as soil, rocks and the materials carrying radon such as water, natural gas, etc.

2.6.1 Radon in Soil and Rock

The original radon comes from the direct decay of ^{226}Ra, which is one of the nuclides formed in the disintegration series from ^{238}U [102].

Therefore, the primarily formed radon in rocks and soil depends on radium concentration and subsequently on uranium concentration in rocks and soils.

Since uranium is litophile element, it predominantly appears in magmata rocks rich in silica such as granites and syenites, and vulcanic rocks such as rhyolite and Porphyry [103].

On a global scale, it is estimated that 2,400 million curies of radon are released from soil annually [8].

In the Nordic countries the air in the pores of soil, the normal radon content is 4,000–50,000 Bq m^{-3} at a depth of one meter.

In fact, the higher concentration of radon is existing in the soil air where there are elevated radium contents [20], [104]. Concentrations of 100,000–200,000 Bq m^{-3} are common in moraines and gravel within areas with uranium-rich granite.

In areas where the soil contains many fragments of alum shale or material from uranium occurrences, the radon concentration can be considerably greater than 1,000,000 Bq m^{-3} [20].

In Yemen, the normal radon content of the air in the pores of soil is 5.582 KBq m^{-3} – 13.56 KBq m^{-3}at a depth of 50 to 150 cm [105].

Freed sandy soil lets the maximum diffusion of radon gas, whereas frozen, compressed or clay soil inhibits its flow. Eventually, it exhales into the atmosphere.

Phosphate rocks contain relatively high concentrations of naturally occurring radioactive materials from the uranium and thorium decay series (^{238}U and ^{232}Th) [106][107].

The concentration of radon in soil gas decreases nearer to the surface as the soil gas escapes to the open air above ground. The concentration of radon in soil gas is affected by meteorological factors, such as barometric pressure, temperature, humidity, wind speed and precipitation [87].

Rocks and soil beneath buildings are normally the main source of indoors radon which is typically four or five times more concentrated than outdoors radon. Radon escapes through the cracks and pores of the soil and rocks in the earth's crust, then it seeps to the indoors air by a variety of factors such as: the difference in air pressure inside and outside the house, cracks, pores, holes in the basement of the house, the type of the soil, soil permeability, soil moisture content, porosity, and the uranium content of soil [108].

Since the soil and the bedrock beneath and/or surrounding the building represent the main source of indoor radon concentration. Therefore, the measurement of the radon concentration in soil can be used as an indicator and a predictive method to evaluate the elevated indoor radon concentrations [109].

2.6.2 Radon in Water

Water quality is one of the most important parameters of environmental studies and it is vital to have regulations about natural radioactivity in drinking water [110].

The radiation doses caused by radon in drinking water are due partly to ingestion but mostly to inhalation of the radon daughters produced by decay of radon released from the water [23]. So, The U.S EPA is concerned with the exposure due to radon in water and is working to set national standards on the amount of radon in water [111]. ^{222}Rn is highly soluble in water. The solubility of radon in water directly varies with pressure and inversely varies with temperature. The concentration of radon in ground water is more than that of surface water [112] because water often moves between the rocks containing uranium and naturally released radon. To compare groundwater radon concentrations in different geologies, aquifers have been classified into two main groups depending on lithology crystalline and sedimentary. The crystalline aquifers contain igneous and metamorphic rocks like granite and gneiss in which ground water flows through fractures.

Water originating from or coming in contact with such granitic or gneissic formations are expected to be associated with elevated levels of radon. In sedimentary aquifers, groundwater flows through interconnected pore spaces, and radon concentration is generally low. Hence, groundwater with elevated levels of radon may be expected to be found in regions with crystalline aquifers, which are generally present in regions where volcanic or orogenic activities have taken place [113].

The highest concentration of radon has been found in granite aquifers [114].

Hess et al. (1983) demonstrated contributions of ^{222}Rn from the water supply to indoor air as high as 36% [115].

Radon can enter the house in a dissolved form through the water supply and be released into the indoor air by such activities as bathing and clothes washing. The concentration in water must be very high to influence the indoor air concentration [57]. A commonly used rule of thumb is that 10,000 pCi L^{-1} (370,000 Bq m^{-3}) of radon in water will produce 1 pCi L^{-1} (37 Bq m^{-3}) when released into the air [57][116].

Water in rivers and reservoirs usually contains very little Radon, because it escapes into the air; so homes that rely on surface water usually do not have a Radon problem from their water. In big cities, water processing in large municipal systems aerates the water, which allows Radon to escape, and also delays the use of water until most of the remaining Radon has decayed [117].

2.6.3 Radon Building Materials

The knowledge of the natural radioactivity of building materials is important for the determination of population exposure to radiations, as most of the residents spend about 80% of their time indoors [118].

Actually, materials used in the typical building may contain significant levels of radium as well as other natural radioactive isotopes of uranium and thorium. Considering the high porosity of construction materials this may cause rather exhalation rate of ^{222}Rn and its accumulation in air of dwellings. Thus, the building materials can represent an important source of indoor radon activity [119], [120].

The exposure rate depends on the following factors: the concentration of radionuclides in the building materials, the amount of building materials used and the type of dwelling. In addition, the surface finish of the building material is important, probably because the surface finish is related to the effective emitting surface, which is larger for an unpolished material [121].

The building industry requires large quantities of low-cost raw materials, and there is an increasing interest in industrial and extractive wastes as substitutes for natural products. This practice may conserve natural resources and cut overall costs, but it may also cause radiological concern [122].

For example, in spite of the enhanced level of radioactivity, phosphogypsum from the phosphate fertilizer industry, fly ash from coal-fired power plants, oil shale ash, alum shale, and some rare minerals containing radioactivity are often used in construction industries. These construction materials may cause radioactivity to be significantly higher than background levels [121].

Another source of elevated radiation comes from the use of granite tiles as decorative material in buildings [106][118] or crushed and mixed with cement to form a brick of standard size [123].

Reported ^{222}Rn emanation factor from several countries had shown that in building materials, it varied from 0.2% to 30%. Building materials baked under high temperature like clay bricks, vitrified tiles and glazed tiles had very low ^{222}Rn emanation factor varying from 0.05% to 0.2% [124].

The study of radioactivity of building materials is required in setting the standards and guidelines for their safe usage and in assessing the radiation hazard associated with them.

Unnecessary exposure to ionizing radiation is considered an undesirable health risk[121].

2.7 Applications of Radon Studies

2.7.1 Radon and Earthquake

Radon is practically an inert gas, its concentration changes are a subject of geological interest disturbances. Sudden disturbance within the earth is manifested at the surface by ground shaking caused by seismic waves [125].

Probably the first to mention radon anomalies associated with earthquakes was Shiratoi in1927. A threefold radon increase in deep ground water observed before the Tashkent(M 5.5) earthquake of 26 April 1966 stimulated the international research on hydro-geochemical precursors [126].

Since then, the occurrence of anomalous temporal changes of radon concentrations in soil-gas, groundwater, hot springs, atmosphere were studied by various researchers [127][128][129][130][131][132][133].

Tectonic deformation during earthquake preparation phase causes changes in rock pressures and fluid convective flows, which lead to changes in the strain field of rock mass.

The strain developed within the earth crust before an earthquake causes transportation of gases from deep inside the earth to the surface anomalous radon emission from soil may be induced due to an increase in crustal compression that squeezes out the soil gas into the atmosphere [134].

Where a slight compression of pore volume causes gas to flow out of the soil resulting to an increase in radon level. Similarly, when pore volume increases, gas flows into the soil from the atmosphere. Thus, an increased radon concentration occurs in the region of compression and radon concentration decreases in the region of dilation. As small changes in gas flow velocity causes significant change in radon concentration, soil radon monitoring is thus an important way to detect the changes in compression or dilation associated with an earthquake event [135].

Correlations between radon anomalies in the ground and earthquakes have raised expectations that radon can be used to help predict earthquakes [64].

2.7.2 Geological Radon

As a geological tool, radon monitoring technique can be used in uranium exploration [136] and hydrocarbon prediction, study of active geological faults and geothermal energy sources. With this technique fault zones have been recognized with a fairly good precision worldwide. The technique can also be effectively used in hydrologic research, when studying the interactions between groundwater, streams, and rivers. It has found limited use in geothermal prospecting [135].

2.8 Risks of Radon Exposure

The first recorded awareness of the effects of environmental radiation on humans was reported by Agricola for miners in the Erz Mountains of eastern Europe in 1556 [63].

In 1879, two German physicians, Hartung and Hesse, pointed out that most of the Schneeberg mine deaths were lung cancers. The Schneeberg miners who had worked in the mines for more than ten years developed the Erz Mountain disease, called bergkrankheit, which was variously diagnosed over the centuries as lymphosarcoma, tuberculosis, malignant tumor of the lungs, bronchogenic carcinoma, squamous-cell carcinoma, and small-cell carcinoma. The diagnosis of bergkrankheit was revised in 1922 to bronchogenic carcinoma [57].

During the period from 1924-1932, it was suggested that radon was the cause of high lung cancer incidence. In 1951, researchers at the university of Rochester N.Y. pointed out that the lung cancer health hazard was from the densely ionizing alpha radiation dose delivered by the radon decay products (^{218}Po and ^{214}Po)that deposited in the respiratory tract [137].

The alpha-particles emitted from radon and its progeny can deliver enough energy to disrupt the DNA and thus cause cancer (as shown in Fig. 2-10. and Fig. 2-11).

Cancers are believed to be caused by the disruption of deoxyribonucleic acid (DNA), which governs the multiplication of cells in the human body. When the DNA's central-control mechanisms fail, cells may grow at an uncontrolled rate. Because each cell splits into two as it develops, the rate of growth of these cells is exponential. If there is no defence mechanism to counteract this effect, a tumour is formed [58].

There is no known threshold concentration below which radon exposure presents no risk. Even low concentrations of radon can result in a small increase in the risk of lung cancer [138].

This is due to the fact that even a single alpha particle can cause major genetic mutation in cell, which may occur at any level of exposure [139].

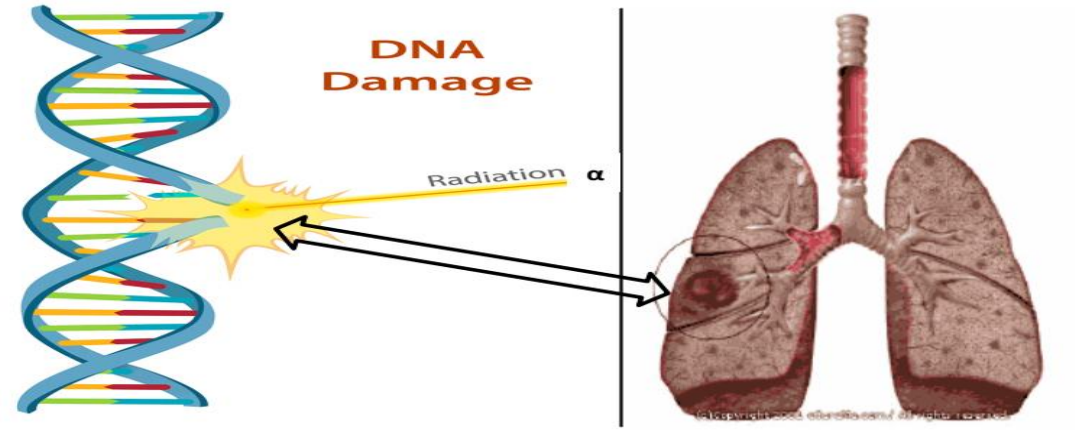

Fig.2-10: Radiation may damage human DNA directly

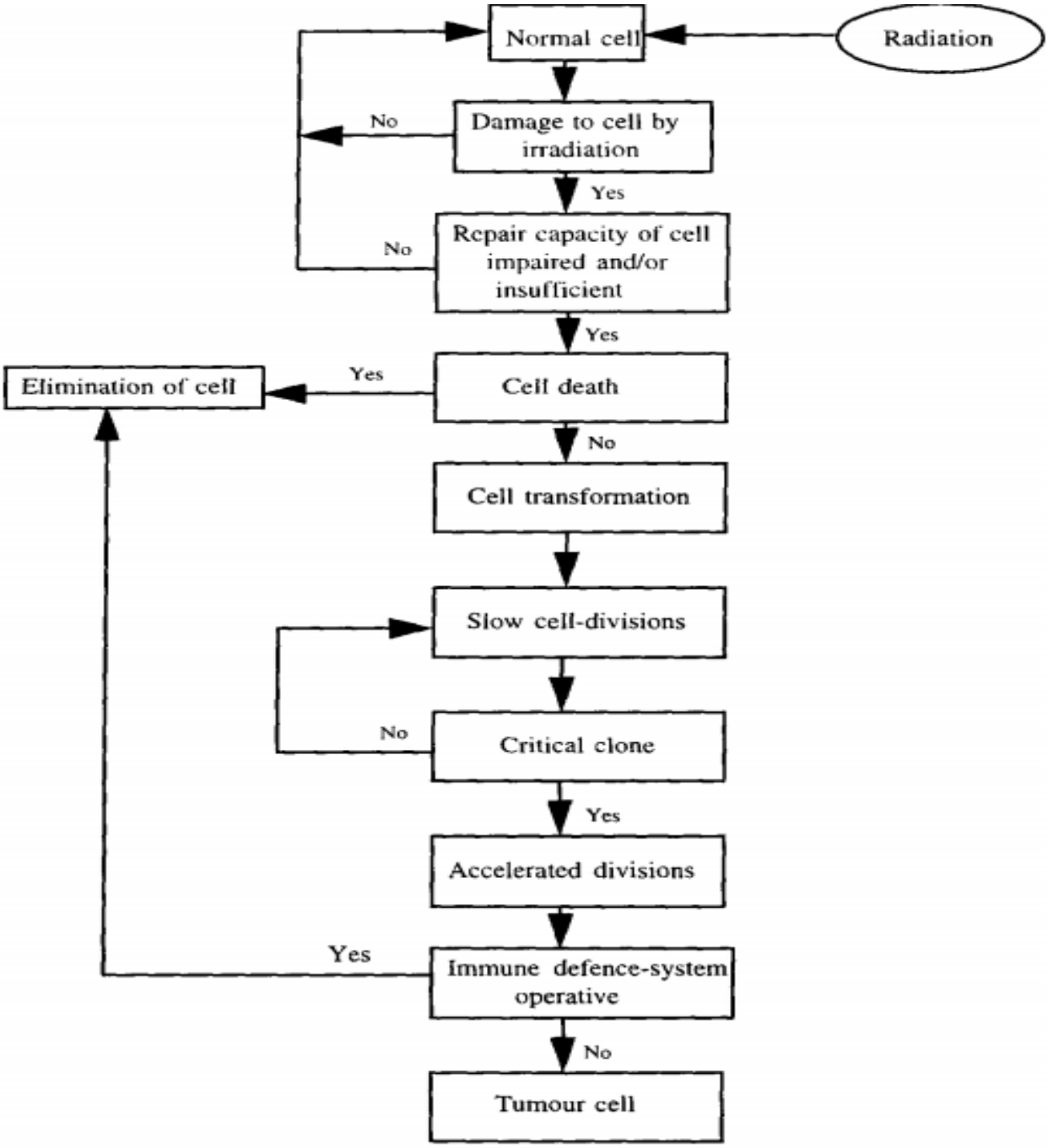

Fig.2-11:Schematic flow-chart showing the sequence of events for cancer induction to occur due to human exposure to radiation [58]**.**

United States Environmental Protection Agency (US EPA) classifies radon as a known human carcinogen on the basis of data from epidemiologic studies of underground miners. That classification is supported by a consensus of national and international organizations [47].

In non-mining contexts, the accumulation of radon in domestic buildings was first observed in 1971 and ascribed to the use of uranium tailings as local landfill [10].

Since then the potential for hazardous exposure due to accumulation of radon in indoor environments became gradually evident, particularly in well-insulated ones [11]. While obvious sources of radiation exposure, such as related to nuclear facilities, are carefully monitored and controlled, radon monitoring in dwellings is scarce and scattered [61].

In 1990, Schoenberg reported that there is a significant relationship between lung cancer cases and indoor radon. Worldwide studies on lung cancer cases have reported that nearly 3–20% of lung cancer deaths are due to indoor radon exposure [140].

Since 1998, the epidemiological study conducted in Iowa US, showed beyond any reasonable doubt that radon decay products cause lung cancer among women who lived at least twenty years in their homes [137].

Canadian statistics show that most Canadians spend ~70% of their time indoors at home, 20% indoors away from home and 10% outdoors. Due to relatively higher radon concentration in residential homes and longer time spent indoors at home, the exposure at home contributes to 90% of the radon-induced lung-cancer risk [141].

There is a well-established synergy between lung cancer from smoking and radon exposure. Smokers and more recent ex-smokers have a much higher baseline risk of lung cancer than lifelong non-smokers. Radon exposure increases this risk still further [142].

The cumulative risk of lung cancer up to 75 years of age is estimated for lifelong non-smokers as 0.4%, 0.5%, and 0.7% for radon activity concentrations of 0, 100, and 400 Bq m^{-3}, respectively. The cumulative risks of lung cancer by 75 years of age for lifelong smokers are close to 10%, 12%, and 16% for radon activity concentrations of 0, 100, and 400 Bq m^{-3}, respectively [138].

Some epidemiological and geographical studies have suggested an association between α–particle exposure via radon and the occurrence of leukaemia, brain tumours, and kidney cancer, especially in children [143].

As some ecologic study suggest that radon may also be a risk factor for skin cancer [144].

Electric field effects can mediate increased exposure to radon daughter nuclei, a known carcinogen [145][143].

In the UK within 400 m of powerlines, this may result annually in 200–400 excess cases of lung cancer, 2000–3000 cases of other illnesses associated with air population and 2–

6 cases of childhood leukemia. Seventeen cases of non-melanoma skin cancer might occur by exposure directly under powerlines [146].

2.8.1 Radon Therapy

For treatment purposes, radon is commonly applied by bathing for about 20 min in water with a radon concentration of 0.3–3 KBq/L or staying for about 1 hour in caves or galleries with natural radon concentrations of about 30–160 KBq m^{-3} [147]. Radon therapy, which mainly emits α-rays and induces a small amount of active oxygen in the body, is performed for various diseases such as osteoarthritis [148].

The findings suggest that radon therapy contributes more to the prevention of life-style-related diseases associated with peroxidation reactions and immune suppression than to thermal therapy. Moreover, these indicate what may be part of the mechanism for the alleviation of hypertension, osteoarthritis (pain), and diabetes mellitus brought about more by radon therapy than by thermal therapy [149].

In fact, radon helped to prevent serious side effects of the pharmaceutical treatment of painful rheumatic and arthritic diseases. While it is estimated that, for example, approximately 12,000 persons annually die from side effects of non-steroid antirheumatic drugs such as diclofenac (mostly due to stomach problems and internal bleeding) in the USA, and more than 1,000 in Germany, no lethal complications have ever been observed from radon treatments.

In addition, it also enjoys an advantage of being inexpensively available in poorer parts of the world with no affordable access to commercial pharmaceuticals for a large part of the population [150].

2.9 Radon Gas Measuring Devices

Radon gas measuring devices can be classified as passive and active devices [151]. Passive devices do not require electrical power or a pump to work in the sampling setting, whereas active devices require electricity and include the ability to chart the concentration and fluctuations of radon gas during the measurement period [152].

2.9.1 Solid State Nuclear Track Detectors (SSNTDs)

Solid state nuclear track detectors (SSNTDs), bare or placed in special chambers (passive dosimeter)have been used for a long time for radon measurements [153], see Fig. 2-12.

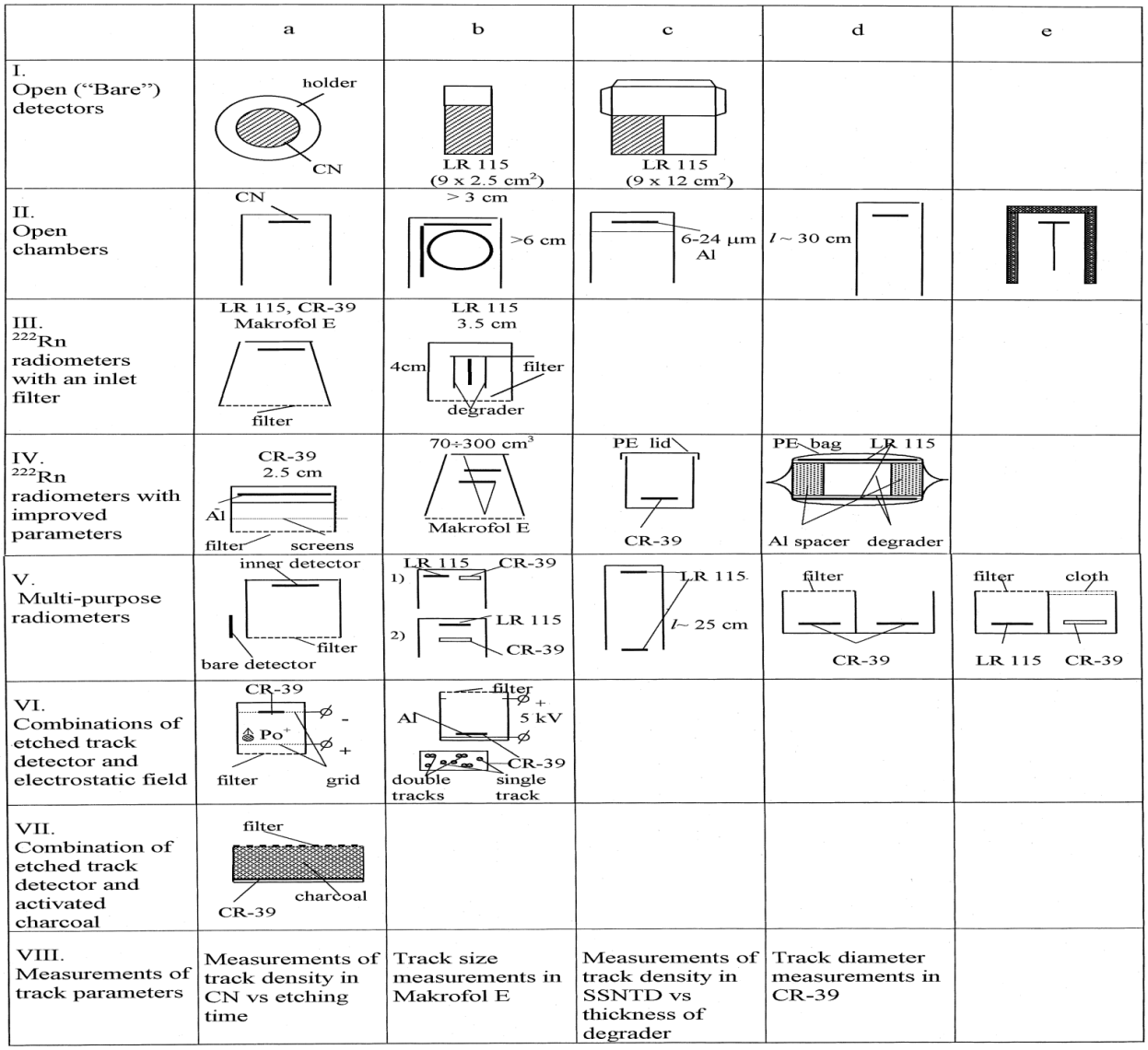

Fig.2- 12: Range of devices for radon measurement based on utilization of solid state nuclear track detectors (SSNTDs). The length of the tube is denoted by l, while abbreviations PE and CN stand for polyethylene and cellulose nitrate, respectively[99].

Also called alpha-track detectors and are based on a detecting material that is impacted by alpha particles, which produce microscopic areas of damage called latentalpha tracks. After the exposure period, such areas are counted either manually or automatically in a

laboratory. The total number of tracks is proportional to the integrated radon concentration [154].

The most frequently applied detectors are cellulose nitrate(LR 115) and delayed neutron counting (DNC), polycarbonate (Makrofol E), and allyldiglycol carbonate (CR-39) [99].

Fig. 2-13 shows radon gas measuring devices: LR 115 and CR-39 detectors.Solid state nuclear track detectors are small, cheap, simple, non-toxic and non-hazardous. They can be sent through the post with instructions for their placement and return [142].

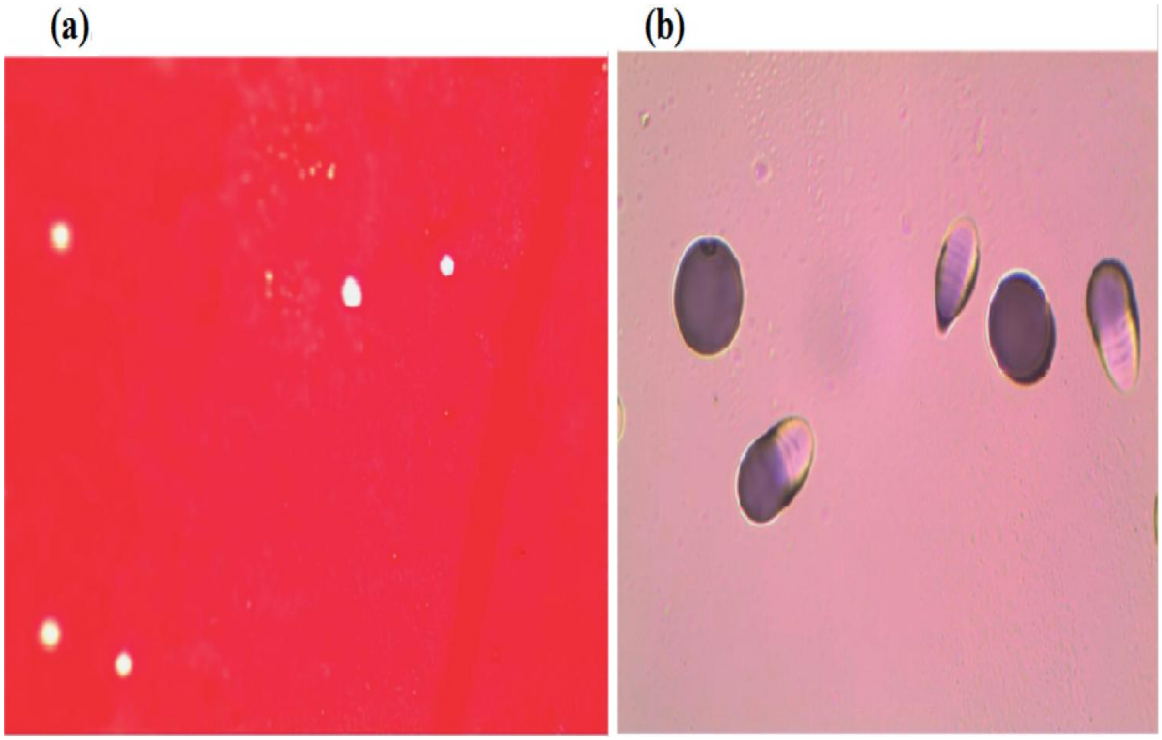

Fig.2- 13: Radon gas measuring devices : (a) LR 115 and(b) CR-39 [155].

2.9.2 Activated Charcoal

In fact, in the early part of the 20th century, Rutherford [156][157] made use of the ability of activated charcoal to adsorb ^{222}Rn to control laboratory contamination [158].

Activated carbon in canisters (See Fig. 2- 4) is now widely used and is recommended for indoor radon measurements by the United States Environmental Protection Agency (US EPA), as well as by numerous other public agencies and private companies. The canister, which is impervious to radon diffusion, is exposed to indoor air which contains radon for a specified period of time. During this time period, radon is allowed to adsorb passively onto the surface of the charcoal [159].

The collector with the adsorbed radon on the carbon is sealed after exposure and returned for analysis to a central laboratory. Counting is usually performed after a minimum of 3 hours have elapsed from the end of exposure to allow equilibration between radon and its progeny. The analysis is straightforward and utilizes equipment that is very common to most radiological laboratories. There are two different detection methods used: (1) gamma counting which is usually done on a NaI (T1) system with a multichannel pulse height analyzer; or (2) alpha and beta counting in a liquid scintillation system [160].

Radon adsorption coefficient, which is defined as the ratio of radon activity on charcoal to radon concentration in air at equilibrium, is commonly used to describe the adsorption ability of activated charcoal. Radon adsorption coefficient not only depends on the properties of the activated charcoal (porosity, grain size), but it is also influenced by environmental factors such as air pressure [161], temperature, humidity [159] and carbon dioxide [162] which have been found to have an adverse effect on the adsorption coefficient [162][159].

Fig.2-14: Radon gas measuring devices: Activated Charcoal [163]

2.9.3 Electret Ion Chamber (EIC)

The EIC radon monitor consists of an electrically conducting chamber having an electret (electrically charged and processed piece of Teflon, exhibiting a stable charge) at the bottom and a filtered inlet at the top as shown in Fig. 2-15. The electret collects ions generated by the radon and the associated decay products inside the chamber [164].

The electret serves a dual purpose: a source of an electric field and as a quantitative sensor. The ions formed inside the chamber during the exposure cause the surface voltage of the electret to drop in proportion to the integrated radon concentration. The voltage drop is determined from measurements before and after exposure [160].

The electret discharge in volts is measured using a noncontact battery-operated electret reader. This value, in conjunction with a duration and calibration factor, yields the radon concentration in desired units. Typical short-term EICs are designed to measure radon for 2 to 15 days at a concentration of 150 Bq m^{-3}. The long-term EICs measure radon over 3 to 12 months at a concentration of 150 Bq m^{-3} [165][152]. Electret ionization chambers need to be handled with care, as dropping them or touching the electret's sensitive surface can cause a partial or total discharge, and thus an overestimation of the concentration of ^{222}Rn. Electret ionization chambers are also sensitive to environmental conditions such as air pressure or a high level of air humidity leading to water condensation on the internal surfaces. In principle, they are not too large, but their sensitivity, in particular to mechanical shocks, would reduce their practicability. Electrets are usually used for screening and diagnostic measurements. The detectors are not usually used for large scale surveys [142].

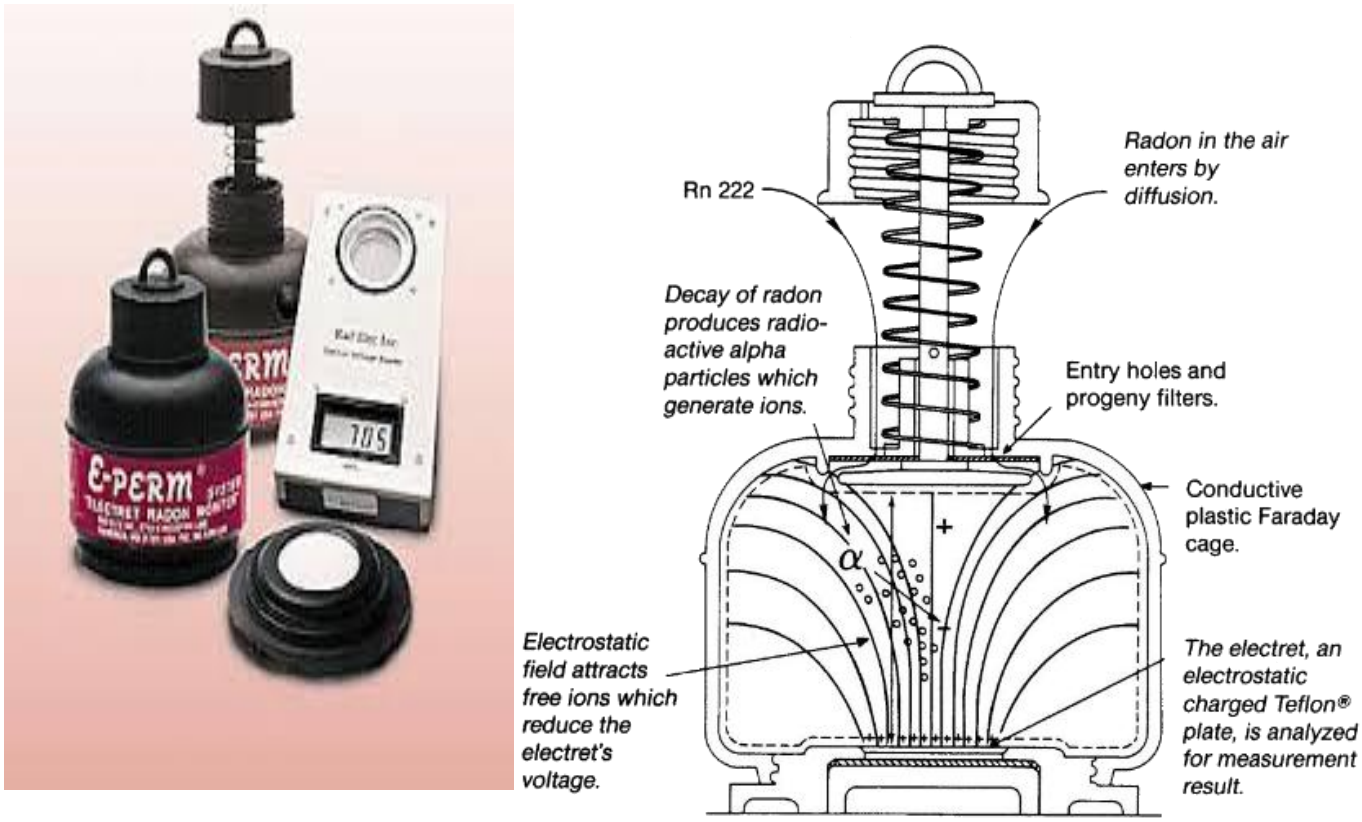

Fig.2- 15: Radon gas measuring devices: Electret Ion Chamber [166][167]**.**

2.9.4 Electronic Integrating Devices (EIDs)

Most electronic integrating devices are able to count the alpha particles emitted by radon and its decay products by making use of a solid-state silicon detector and a diffusion chamber. These devices have two main restrictions. First, they usually require long integration periods (more than two days), because of the small dimensions of their diffusion chambers. Second, their measurements can be altered in high humidity environments [154]. Higher sensitivities can be achieved by applying high voltage to collect the charged radon decay products electrostatically by direct contact to the detector. High air humidity may affect the measurement [152].

2.9.5 Continuous Radon Monitors (CRMs)

There are three types of continuous radon monitors (CRMs) .In the first type, the continuous radon monitor designed for field deployment includes a scintillation probe, a counting and printing unit, and a power-supply unit. The scintillation probe includes a cylindrical cell which is coated with activated ZnS in the inner wall for alpha scintillation, a photomultiplier tube, a preamplifier, and a high-voltage power supply [168].

A second type of CRMs operates as an ionization chamber. Radon in the ambient air diffuses into the chamber through a filtered area so that the radon concentration in the chamber follows the radon concentration in the ambient air with some small time lag. The third type of CRMs functions by allowing ambient air to diffuse through a filter into a detection chamber. As radon decays, the alpha particles are counted using a solid state silicon detector [169].

If very accurate measurements, particularly of very low radon concentrations, are necessary, then a pulse ionization chamber is likely to be the instrument of choice. Such equipment is expensive and complex and it is generally not practicable for large scale use [142].

Some devices eliminate the cross-sensitivity to air humidity by drying the incoming air. CRMs require routine calibrations to assure proper functioning and reliable results [152]. Figure 2-16 shows some types of continuous radon monitors.

Fig.2- 16: Radon gas measuring devices: continuous radon monitors [103][152]**.**

Some of the advantages of doing a continuous radon monitor test are:

The continuous radon monitor has the ability to time integrate the radon measurement. Most continuous radon monitors, as a minimum, integrate hourly, but some higher end models can be set to integrate anywhere from 1 minute to 1 week. Some continuous radon monitors have a digital readout that can be set to display the current radon level or the long term average radon level. Most continuous radon monitors have the ability to collect and store their measurement data [170].

Some disadvantages of a continuous radon monitors are being expensive and complex and it is generally not practicable for large scale use. Measurement systems using active sampling are expensive in terms of staff time and data analysis and for this reason are usually used only in diagnostic or research applications [142].

Many models of continuous radon monitors come equipped with other environmental sensors to simultaneously measure other parameters like; barometric pressure, ambient temperature and relative humidity [171].

2.9.5.1 The Lucas Scintillation Cell (LSC)

The Lucas scintillation cell (LSC) is commonly used all over the world for the estimation of radon. The cell was originally devised by Vandilla and Taysum (1955) [172].

The (LSC) comprises a ~100 cm^3 or larger vessel with a transparent window. The inner walls of the LC are covered with ZnS(Ag) scintillator a few tens mg cm^{-2} thick. The inner surface of the window is usually covered with a transparent layer of tin oxide to assure conductivity and prevent a buildup of ^{222}Rn daughters on the window [173].

In use, a filtered sample of the air to be measured for radon concentration enters the cylinder. After waiting for radioactive equilibrium between radon and its short-lived alpha-emitting progeny, the Lucas cell is placed on a photomultiplier tube (PMT) inside a light-tight enclosure [174].

The scintillations or flashes of light caused by the alpha particles from radon, ^{218}Po and ^{214}Po which strike the ZnS(Ag) are recorded by the PMT and its associated electronics. Using appropriate calibration and decay scheme factors, the radon gas concentration may be determined from the rate at which the pulses are recorded [175].

The characteristics of radon measuring devices used by the international Radon Project of WHO from 2005 to 2007 are described in Table 2-2.

Table 2- 2: Radon gas measurement devices and their characteristics [152]**.**

Detector Type(Abbreviation)	Passive/Active	Typical Uncertainty[a][%]	Typical Sampling Period	Cost
Alpha-track Detector(ATD)	Passive	10 - 25	1 - 12 months	low
Activated Charcoal Detector (ACD)	Passive	10 - 30	2 - 7 days	low
Electret Ion Chamber(EIC)	Passive	8 - 15	5 days - 1 year	medium
Electronic Integrating Device (EID)	Active	~ 25	2 days - year(s)	medium
Continuous Radon Monitor (CRM)	Active	~ 10	1 hour - year(s)	high
[a]Uncertainty expressed for optimal exposure durations and for exposures ~ 200 Bq/m^3 .				

2.10 Special Quantities and Units for Radon

This section sets out the special quantities and units that are used to characterise the concentration of the short-lived progeny of radon in air, and the resulting inhalation exposure.

• Becquerel

A becquerel (Bq) is a measure of activity equal to one disintegration per second.

One curie (Ci) is equal to 3.7×10^{10} Bq [111].

•The potential alpha energy:

The potential alpha energy of an atom of the decay product is the sum of alpha energies emitted during the decay of this atom up to ^{210}Pb.

• Potential alpha energy concentration:

The PAEC of any mixture of activity concentrations of short-lived ^{222}Rn decay products is the sum of the potential alpha energy of these atoms present per unit volume of air. Therefore, for determination of the PAEC in air, the activity concentration of individual ^{222}Rn decay products has to be known.(symbol PEAC units MeV/l or J/m^3) [176]**.**

•Equilibrium equivalent concentration:

Equilibrium equivalent concentration of radon (EEC), for a non-equilibrium mixture of short-lived Rn daughters, is that (smaller) activity concentration of Rn in secular radioactive equilibrium with its daughter products that has the same potential alpha energy (PAE) concentration.(symbol EEC, units Bq m^{-3}) [177].

• Working level:

One Working Level (WL) equals any combination of radon progeny in 1 L of air, which results in the ultimate emission of 130 000 MeV of energy from alpha particles. WLM is a time-integrated exposure measure and is the product of time, in units of working months, which is taken to be 170 hours, and WL. In terms of SI units, 1 WLM corresponds to 2.08×10^{-5} (J m^{-3}) × 170 hours 4 3.5×10^{-3} J h m^{-3} [178].

• Potential alpha energy exposure:

Given that the 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~170 h per month) [179].

Chapter 3: The Experimental Work

3. The experimental Work

3.1 The Experimental Work Stages in Building Materials

3.1.2 Objective of the Work

The aim of this study is to determine the annual effective doses of ^{222}Rn exposure, effective radium content and radon exhalation rates in some building materials from the local market of Ibb province in Yemen and Rabat, the capital city of Morocco, by solid-state nuclear track detectors (CR-39).

The Findings obtained in this research can be used in various ways such as:

I. Formulation of policies regarding public health and the environment.
II. Raising public awareness on the possible consequences of radiation exposure.
III. Such studies can be useful for making reference records and developing data panels to ascertain changes in the environmental radioactivity due to nuclear, industrial and other human activities over time [180].

3.1.3 Sampling of Yemen

For the purposes of this study, a total of 33 samples of building materials were collected from different companies and stores in Ibb Province, comprising 15 slab samples and 18 porous powdery building material samples. Ibb Province is located between Thamar and Taiz provinces. The capital city of Ibb province is located some 193 km to the south of Sana'a, the capital city of Yemen. Fig.3-1 shows Ibb's location [181]. The province has a total area of 5383 km^2 and it is located at latitude 13°58′48″ and longitude 44°10′48″[182].

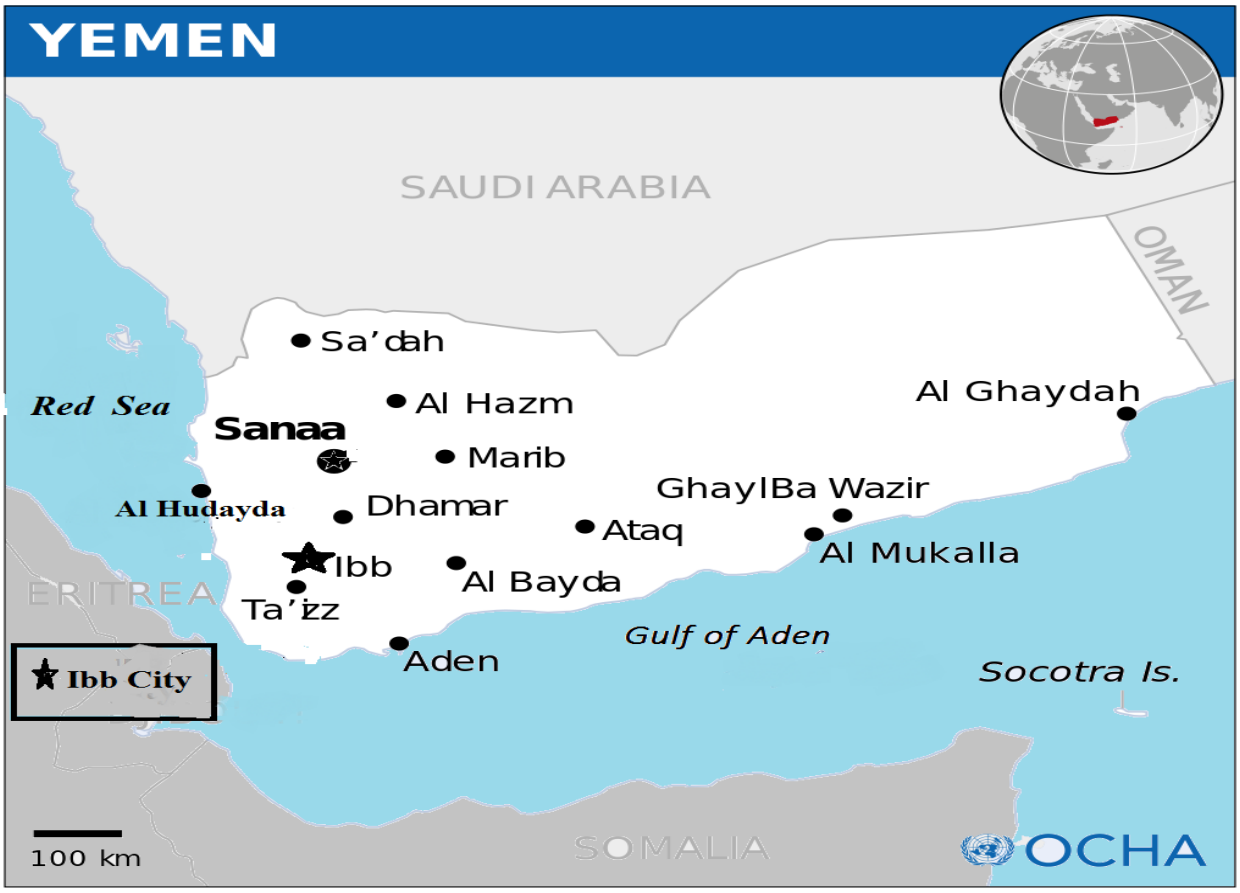

Fig. 3- 1: Map showing the study area in Ibb [183]**.**

3.1.4 Sampling of Morocco

This study has also been conducted in Rabat, the capital of Morocco. Fig.3-2 shows Rabat's location [89]. The study area stretches over 70 Km^2. The geographic coordinates of Rabat, Morocco are: latitude: 34°1 N, longitude: 6º50 W [184].

A total of 35 samples of building materials were collected from different stores and companies in Rabat, comprising 19 slab samples, and 16 porous powdery building material samples.

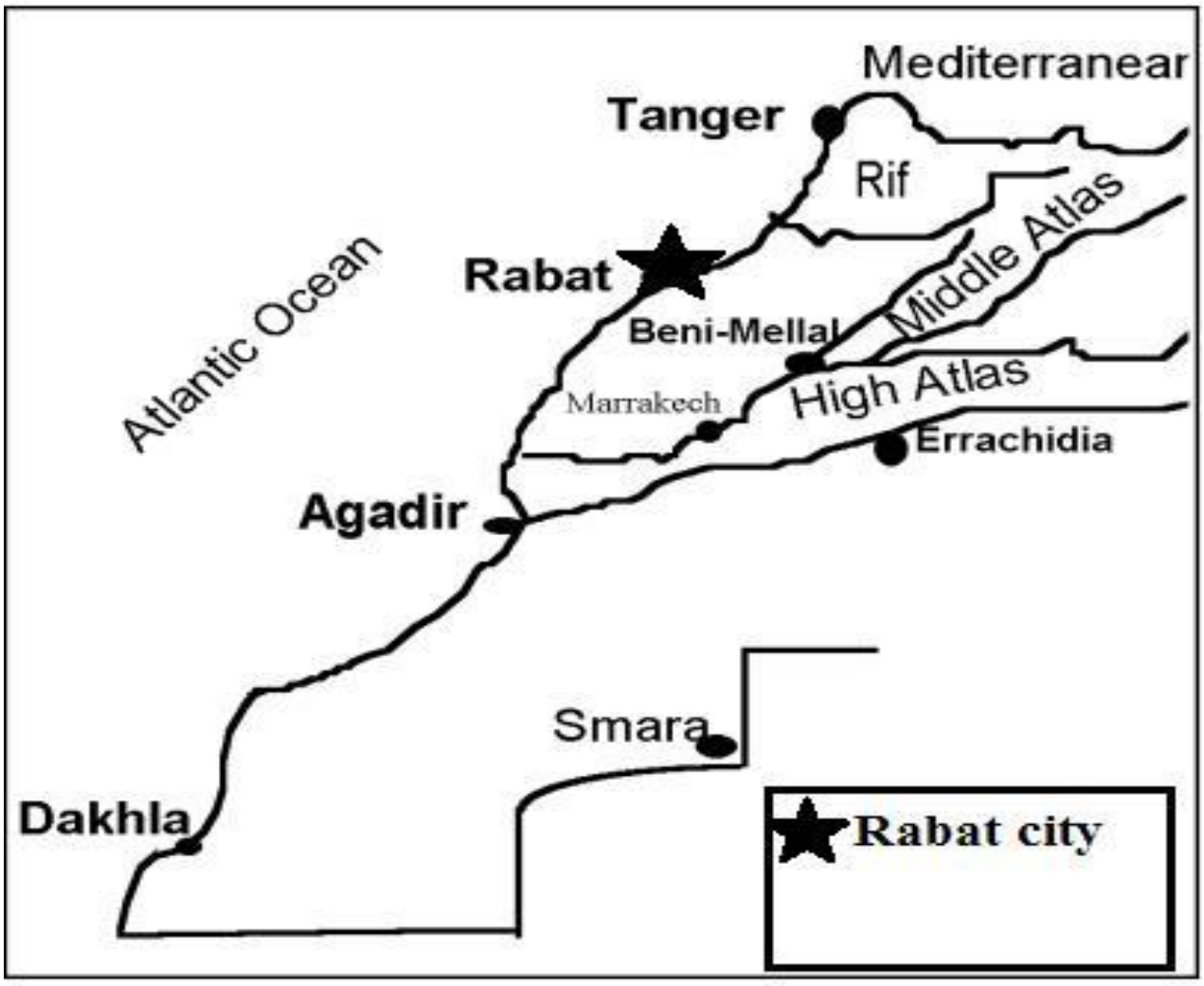

Fig. 3- 2: Map showing the location of the studied samples in the Rabat city, Morocco[89]**.**

3.1.5 Preparation of Dosimeters

CR-39 detectors have a good capability to register tracks at different levels of registration sensitivity. They are also used to measure the concentration and exhalation rate of radon [92], [185], [186]. The CR-39 polymer sheets of TASTRAK were produced and provided by Track Analysis Systems Ltd. (TASL), Bristol, UK. The polymeric detector samples, for the present study, were cut to a size of 1.5×1.5 cm^2 and adhered to a plastic can of known dimension (7 cm in diameter and 11 cm in height) [187]–[190]. In order to avoid the track contribution from thoron in the can, the CR-39 detectors were kept at distance approximately 7 cm or more from the sample in accordance with the protocol [191]–[195]. This is because the half-life time of ^{220}Rn (55.6 s) is about 0.00017 that of radon ^{222}Rn (3.82 d). This shows that most of the thoron will decay inside the building material and radon diffusion length, so that, thoron would be still in the order of 2 or 3 cm [95], [196], [197].

3.1.6 Preparation of Samples

In this study, we used two different types of experiments in the measurement process of radon exhalation rates of building materials:

1-The arrangements of our experimental tools are started by placing the containers inversely on the surface of the tile samples of building materials. These samples were washed, cleaned, dried and used without crushing as used naturally. The contact between the chamber and the building material parts was sealed with silicon [123] as shown in Fig.3-3.

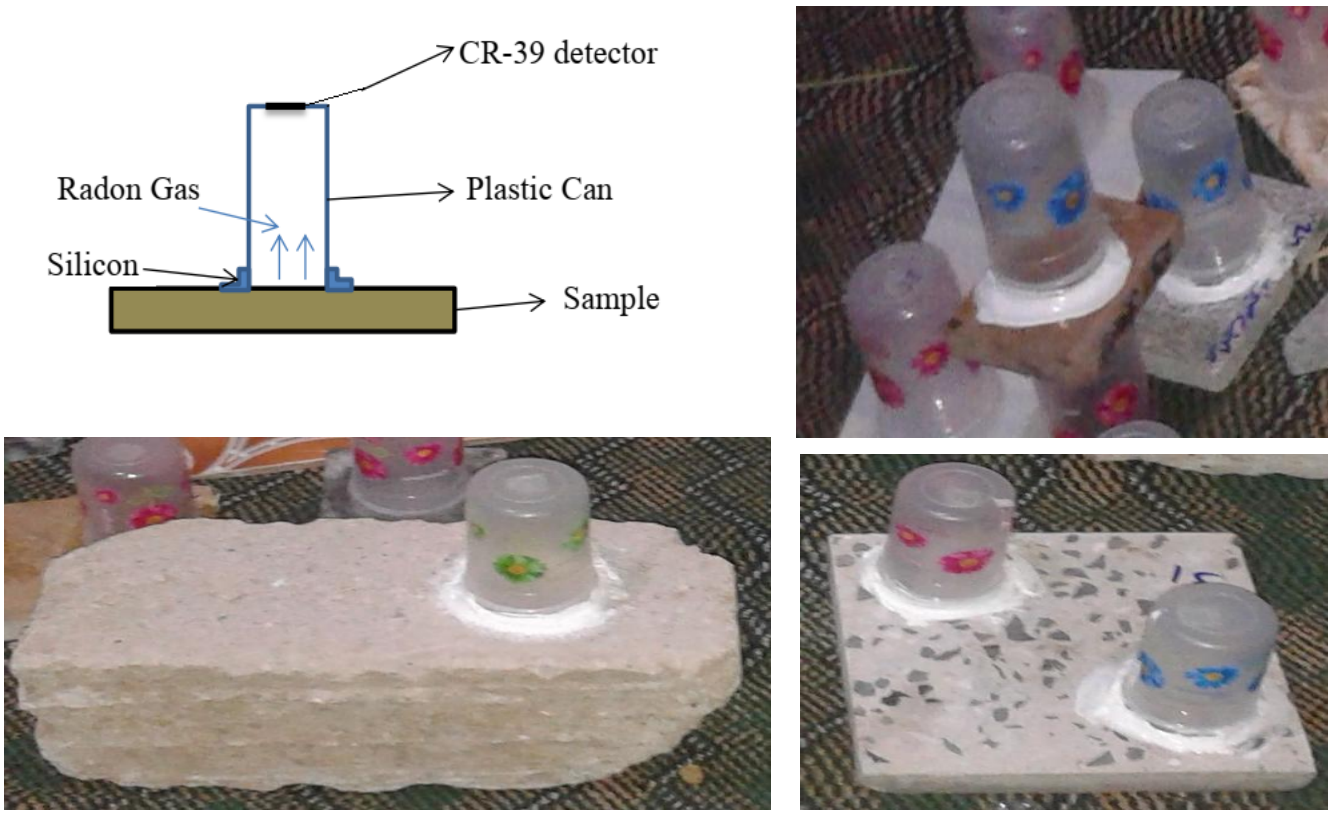

Fig. 3- 3: Experimental set up for the measurement of radon exhalation rate.

2- In the second part, were moved all the moisture content from our powder samples by dried them in an oven at a temperature of 80°C for 24 h [186], [198] and crushed to fine grain size (100 μm) [199]. Then each sample of 125 g weight was placed at the bottom of a cylindrical sealed can as shown in Fig.3-4. The mouth of the cylindrical can was sealed with a cover and fitted with CR-39 plastic track detectors at the top inner surface so that they were facing the specimen [200]–[202]. The cover was sealed by silicon and duct tape from the outside in order to avoid radon leakage [203]. The sample was left for one month to allow radioactive equilibrium to reach between ^{226}Ra,^{222}Rn and their decay products. Furthermore,

in order to ensure the radioactive equilibrium of ^{226}Ra, ^{222}Rn and their decay products, it is important to ensure that no ^{222}Rn is lost from the sample container [199], [204]. During the exposure period of 3 months [205], [206], the sensitive side of the detector is exposed freely to the emergent radon from the sample in the can so that it could record the tracks of alpha-particles resulting from the decay of radon.

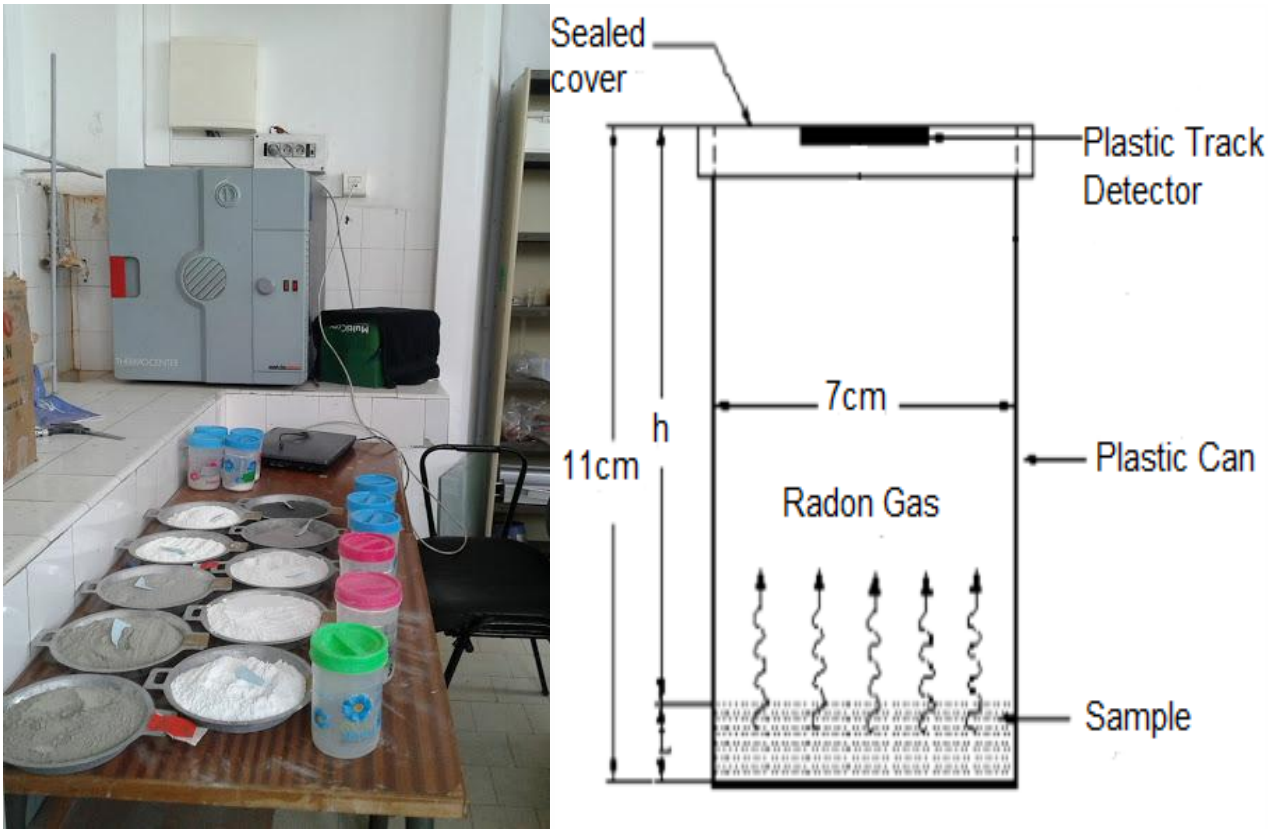

Fig. 3- 4: Experimental set up for the measurement of radon exhalation rate.

3.2 The Experimental Work Stages in Soil and Fertilizers

This section consists of two parts: (a) Measurement of radon concentration in soil, (b) Measurement of radon exhalation rates from fertilizers, where CR-39 detectors have been used in both the measurements.

3.2.1 Part one: Measurement of radon concentration in the soil

Some studies found that soil gas levels measured below 10 KBq m^{-3}could produce indoor radon concentrations above the working level of 200 Bq m^{-3} [152]. And it is known that every 1 Bq m^{-3} of the radon level means about 0.015 mSv y^{-1} [153], [154].

Also, soil is commonly used as building materials for mud houses as shown in Fig.3-5. And the origin of mud material may contain naturally the rare elements without decomposing, and this would contribute to a higher probability of radon concentration. It has been reported that radon levels are higher in mud buildings as compared with the concrete buildings [155].

These studies highlight the importance of measurement of radon concentration in the soil.

Yemen's Qaa Al-Hakel district is an agricultural area and mud constructions are the most common type for building houses in this area. Therefore, the determination of radon concentration in the soil will be of great importance. This will help evaluate and prevent radon-related health hazards in dwellings.

3.2.1.1 The Study Objectives

This study investigates the concentration of radon gas in the soil of Qaa Al-Hakel agricultural area in Ibb province, Yemen. This study was fielded in location A (3589 m^2) and location B (1205 m^2), with a total area of about (4794 m^2). Solid-state nuclear track detectors (SSNTDs), CR-39, are used in this work.

Fig. 3- 5: Homes made of mud in Yemen

3.2.1.2 Distribution of Dosimeters in the Study Locations

During the horizontal study of radon gas concentration in the soil, 94 dosimeters (location A=60, location B=34) were placed equally at depth of 50 cm, as shown in Fig.3-6.a, noticing that the diameter of each hole is about 20 cm. The detectors were distributed as follows:

First: location A

30 dosimeters (D_1, D_2, D_3……D_{30}) were placed in the western part of the location A of the study with 5 m distance separation between the adjacent dosimeters. Dosimeters had been extended at one line from south to north as shown in Fig.3-7.a.

30 dosimeters (D_{31}, D_{32}, D_{33}……D_{60}) were placed in the eastern part of the location A with 5 m distance separation between the adjacent dosimeters. Dosimeters had been extended at one line from south to north as shown in Fig.3-7.a.

Second: location B

17 dosimeters (D_{86}, D_{87}, D_{88}……D_{102}) were placed in the southern part of the study location with 5 m distance separation between the adjacent dosimeters. Dosimeters had been extended at one line from east to west as shown in Fig.3-7.b.

17 dosimeters (D_{103}, D_{104}, D_{105}……D_{119}) were placed in the northern part of the location of the study with 5 m distance separation between the adjacent dosimeters. Dosimeters had been extended from east to west as shown in Fig.3-7.b.

The dosimeters had been left there for 34 days, then they were collected, but the dosimeters, D_3, D_{24} were lost in the western part of the location A; D_{97} was lost in the southern part of the location B, because it was unintentionally broken when taken out..

Radon gas levels were also studied at ground surface (at depth of zero cm) by putting 10 dosimeters in location A (D_{76}, D_{77}, D_{78}……D_{85}) upside down on the ground surface directly, see Fig.3-6.b. These dosimeters were placed in the northern part of the location A next to the fence with 5 m distance separation between the neighboring dosimeters (see Fig.3-7.a). After an exposure period of 30 days, all dosimeters were collected.

Radon gas levels at different depths were also studied. Twenty three dosimeters (location A=15, location B=8), as shown in Fig.3-6.a, were placed at different depths.

First: location A

61-75 dosimeters (D_{61}, D_{62} …….D_{75}) were placed at different depths (10, 15, 20, 25, 30, 35, 40, 45, 50, 60, 70, 80, 90, 120 and 150 cm), respectively, in the central part of the location A, with 7m separation distance between the adjacent dosimeters as shown in Fig.3-7.a. Then they were collected after 34 days.

Second: location B

120-127 dosimeters (D_{120}, D_{121} ……. D_{127}) were put at different depths (10, 20, 30, 40, 50, 60, 70 and 80 cm), respectively, in the central part of the location, with 4m separation distance between the adjacent dosimeters as shown in Fig.3-7.b. Then they were collected after 30 days. All dosimeters were collected, except dosimeter number D_{126} was lost.

Generally, the total number of the dosimeters used in the study was (127), 4 of which were lost. The average loss was (3.149%).

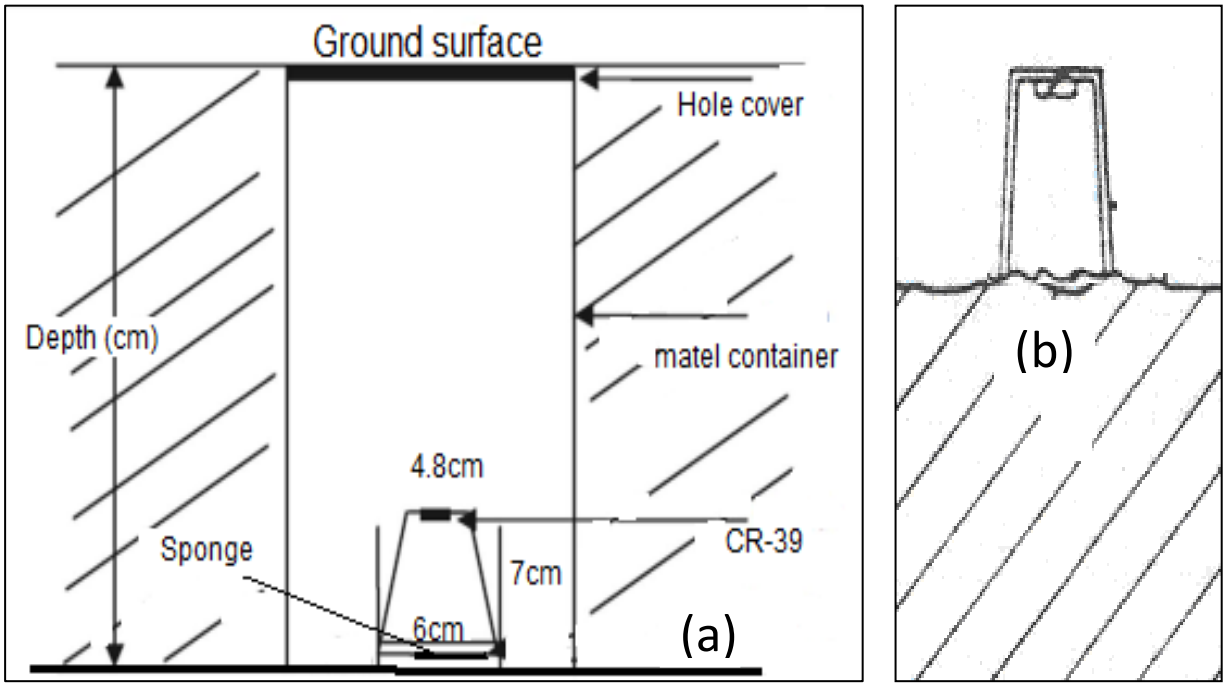

Fig. 3- 6: Radon dosimeters arrangement used in measurement of radon concentration.

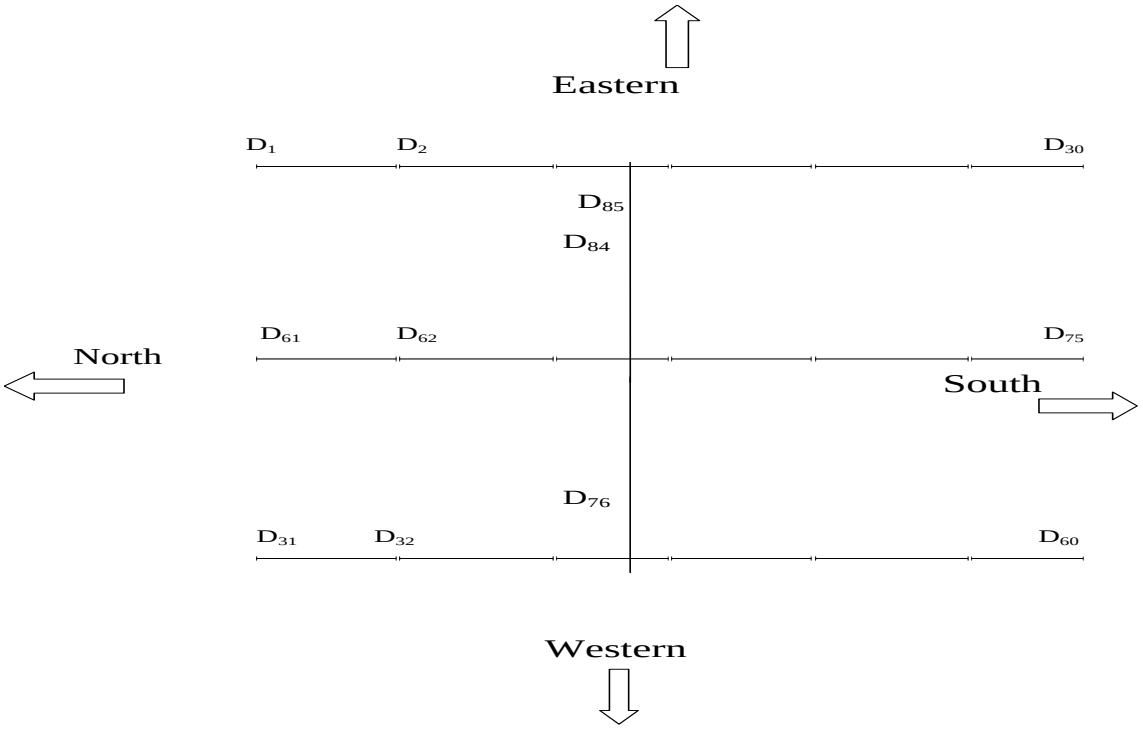

Fig. 3- 7:a. Location A.

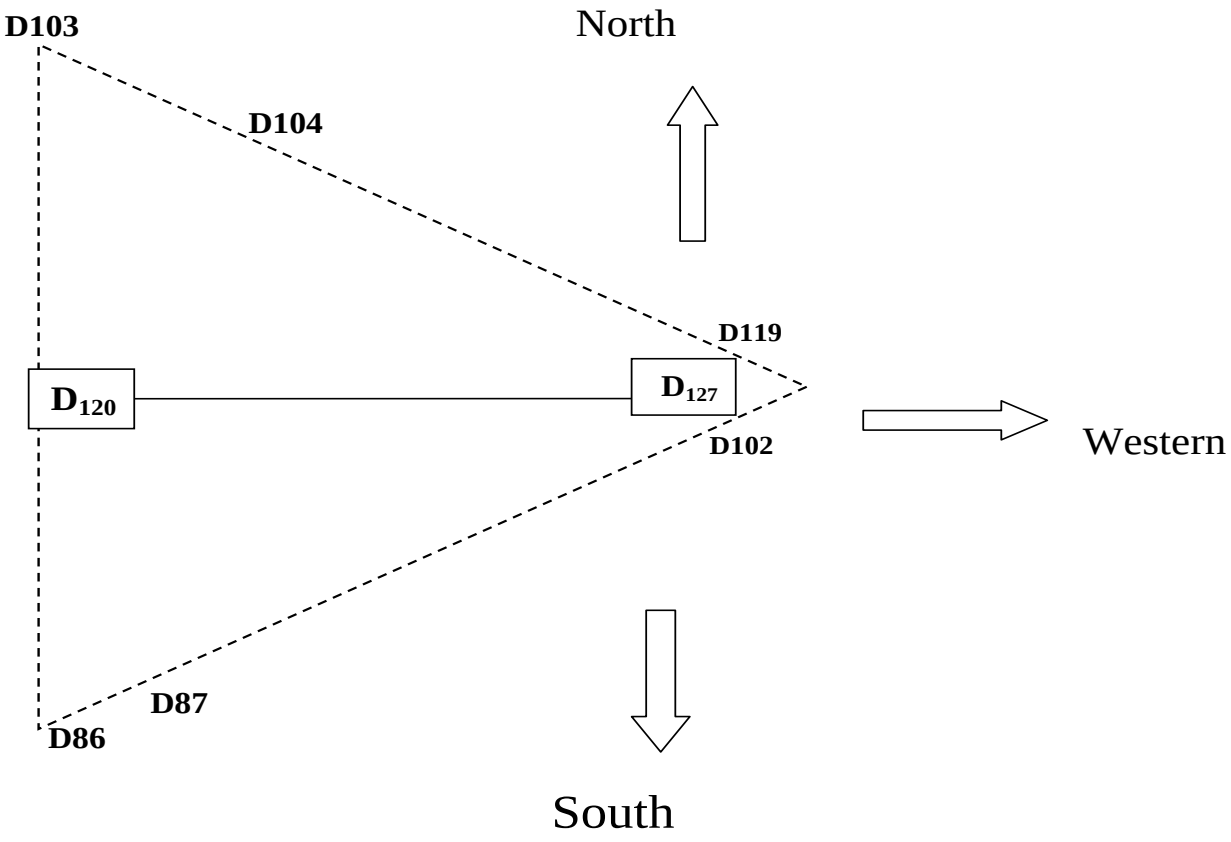

Fig.3-7.b. Location B.

3.2.2 Part Two: Measurement of Radon Exhalation Rates from Fertilizers

Phosphate fertilizers are one of the most commonly used materials in the agricultural field all over the world. The level of activity concentration of radionuclides in phosphate fertilizer and soil samples provides useful information in the monitoring of environmental contamination [27].

In phosphate ores, usually naturally occurring radioactive elements such as ^{238}U, ^{232}Th, etc. and their decay products may exist in high concentrations. The most interesting of these natural radioactive series is the ^{238}U series which decays via a chain containing eight alpha decays and six beta decays to ^{206}Pb. This chain includes the longest-lived isotopes of radium and radon; ^{226}Ra and ^{222}Rn [207].

^{226}Ra has a high potential for causing biological damage through continuous irradiation of human skeleton over many years and may induce bone sarcoma [191].

^{222}Rn is a case of an outcast element in nature because this element is the only gas in this uranium series whereas ^{226}Ra, the first direct parent of radon, is a solid radioactive element and decays to radon (^{222}Rn) by emitting alpha particle [207] as shown in Fig.2-7. Uranium and radium are introduced to surface soil by using N - P - K fertilizers in agriculture in the form of very soluble phosphate complexes.

Groundwater plays an important role in the migration and redistribution of the uranium and radium in the Earth's crust [208].

3.2.2.1 Study Objectives

The aim of this study is to evaluate the concentrations and exhalation rates of radon from different samples of Fertilizers used locally in Yemen using a solid-state nuclear track detector (SSNTD) (CR-39).

3.2.2.2 Materials and Methods

A total of 12 samples of fertilizers were collected from different companies and stores in Ibb city.

Samples were prepared for measurement in the same way of preparing the building material samples (powder samples) for the same exposure time of 3 months.

3.3 Chemical Etching

At the end of the exposure period, All CR-39 detectors were collected and mounted vertically in a stainless steel spring and then immersed in the etching solution 6.25 N NaOH inside a water bath at 70 C for 9 hours.

Therefore, to prepare 1 normal solution, 1 gram mole of NaOH is dissolved in 1 liter of distilled water.

1 molecular mass of NaOH is calculated as follows:

The atomic masses of Na = 23, O = 16 and H = 1.

Thus, the molecular mass of NaOH = (23 + 16 + 1) = 40 gm.

Therefore, 40 gm of NaOH dissolved in 1 liter of distilled water gives 1N solution of NaOH.

Hence, 240 gm of NaOH (6 x 40) dissolved in 1 liter of distilled water gives 6 N solution of NaOH.

At the end of the etching process the detectors were washed thoroughly with distilled water and then left to dry.

The etching process was performed at Chemistry Laboratories of Faculty of Sciences, Mohammed V University using the equipment shown in Fig.3-8 below.

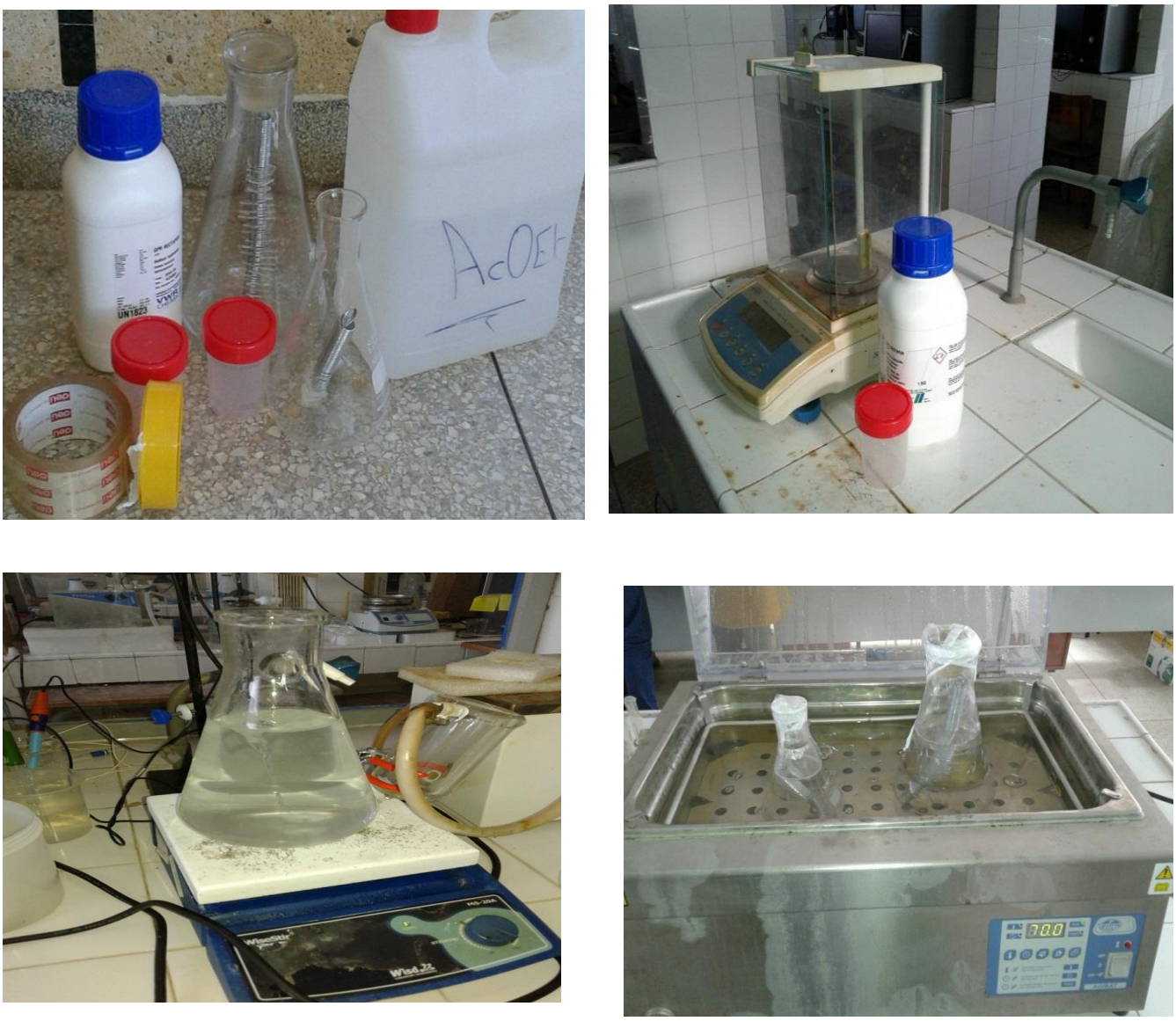

Fig. 3- 8: Equipment used in Etching Process.

3.4 Observation under Optical Microscope

Each detector was counted visually using an optical microscope with a power of (40 x 10). Etch pit "track" sizes and shapes will of course vary: vertically incident alpha particles will form circular etch pits. While the majority of etch pits will be elliptical resulting from alpha particles incident on the detector surface of shallower dip angles.

Then consistently ignore any smaller etch pits and any scratches that are easily discounted. The genuine track etch pit may be identified by slowly moving the fine focus of microscope up and down and looking for a bright point of internally reflected light at bottom tip of the etch-pit. See Fig.3-9.

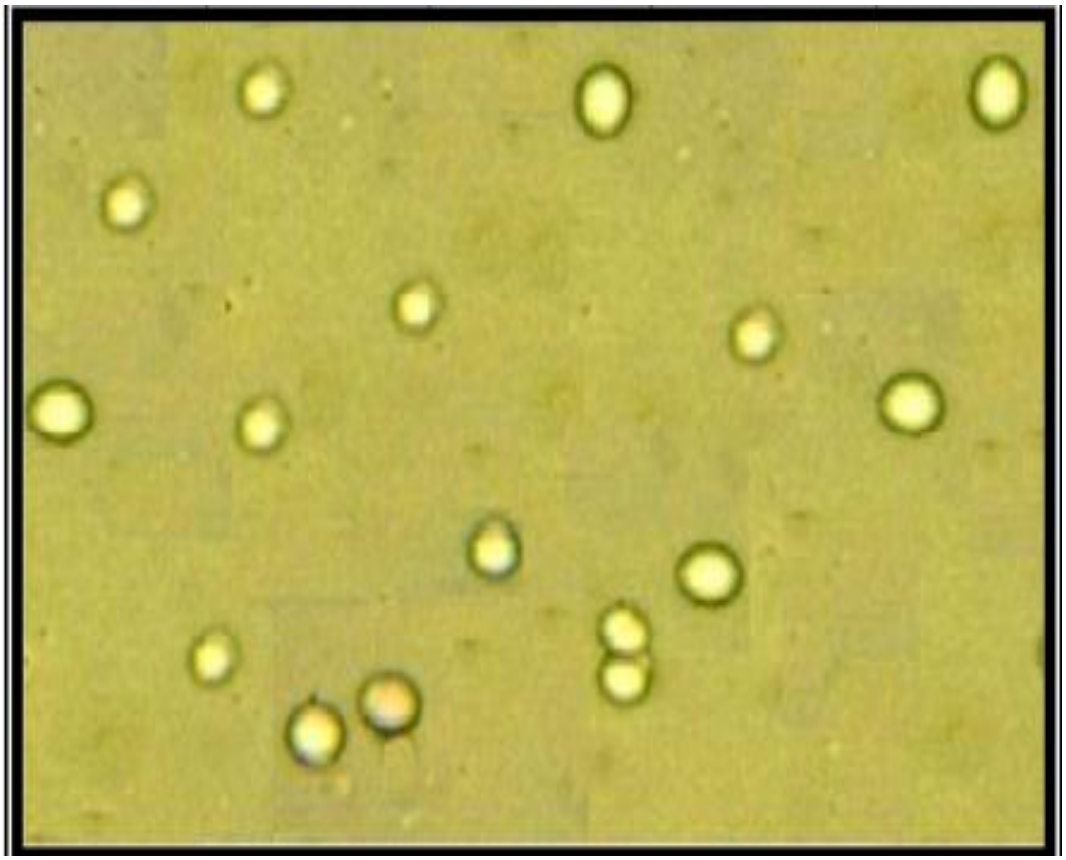

Fig. 3- 9: The tracks in the field view.

3.5 Counting of Tracks Density per Unit Area of the Detectors

In order to calculate the radon exhalation rate, it is necessary to know the track density, the exposure time and the calibration factor that converts the track density to radon exhalation rate.

To count the average tracks $\bar{N}$ found on the detector surface, the pits were read in 50 fields of view for each detector [209]. Then these pits were calculated (N_{tot}) in each field of view and were divided by the number of fields of view read before n=50.

$\therefore$ The average of tracks can be counted by the following equation:

$$\bar{N} = \frac{N_{tot}}{n} \qquad (3.1)$$

Where $\bar{N}$ is average number of tracks, N_{tot} is the total number of tracks, and n is the no. of fields of view read from the detector's surface.

We obtain the density (ρ) from the following equation:

$$\rho = \frac{\bar{N}}{A} \qquad (3.2)$$

where the area of the field of view from the detector's surface, see Table 3-1.

The length and width of field of view were found by slide has a special scale to measure the small dimensions (in μm), see Fig.3-10.

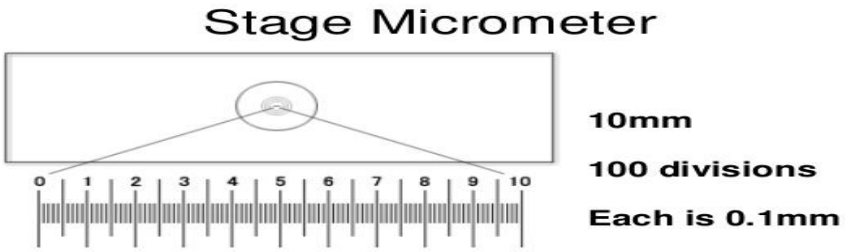

Fig. 3- 10: Stage Micrometer Glass slide

3.6 Radon Concentration in Building Materials

Conversion of alpha track density to radon concentration is based on a factor obtained from calibration of CR-39 detector in a radon chamber of known concentration. Radon concentration per unit volume is calculated using the following equation [210]–[212]**:**

$$C_{Rn} = \frac{\rho}{KT} \qquad (3.3)$$

Where C_{Rn} is the radon concentration (Bq m^{-3}), T is the exposure time (day), ρ is the track density (Tr cm^{-2}), K is a calibration factor determined from the radon chambers which was calibrated in previous works, see Table 3-1.

3.7 Radon Exhalation Rate from Building Materials

The mass exhalation and surface exhalation rate of radon were obtained from the following expressions [190], [206], [213]:

$$E_A = \frac{C\lambda_{\mathrm{Rn}} V}{A[T + (1/\lambda_{\mathrm{Rn}}(e^{-\lambda_{\mathrm{Rn}} T} - 1))]} \qquad (3.4)$$

$$E_M = \frac{C\lambda_{\mathrm{Rn}}V}{M[T + (1/\lambda_{\mathrm{Rn}}\,(e^{-\lambda_{\mathrm{Rn}}T} - 1)\,)]} \qquad (3.5)$$

Where E_A is the surface exhalation rate(Bq $m^{-2}h^{-1}$), E_M is the mass exhalation rate(Bq $kg^{-1}h^{-1}$), C is the integrated radon exposure (Bqm^{-3} h), A is the total surface area of the sample from which radon is exhaled (3.85×10^{-3} m^2), V is the volume of air in the can (m^3) or (effective volume of the can), M is the mass of the powder sample (0.125 kg), $\lambda_{\mathrm{Rn}}$is the decay constant of radon (h^{-1}) and T is the exposure time in hour.

The risk of lung cancer from domestic exposure due to radon and its daughter nuclides can therefore be computed directly from the effective dose equivalents. Actually, we can calculate the radiation hazards due to radon and its daughters from the radon exhalation rates of building material samples. In addition, The contribution of indoor radon concentration from building materials can be calculated from the following relation [214]:

$$\bar{C}_{\mathrm{Rn}} = \frac{E_A \times S}{(\lambda_{\mathrm{Rn}} + \lambda_{\mathrm{V}}) \times V_r} \qquad (3.6)$$

where $\bar{C}_{\mathrm{Rn}}$ is radon concentration in construction materials contributing to indoor radon, V_r is room volume (m^3) and λ_{V} is air exchange rate (h^{-1}). In these calculations, the maximum radon concentration from building materials was assessed by assuming the room to be a cavity with the ratio S/V_r = 2.0 m^{-1}, where S and V_r are the internal surface area and volume of the room, respectively, and the air exchange rate λ_{V} was taken to be 0.5h^{-1} [215], [216].

The annual effective doses of ^{222}Rn exposure were estimated using the the below equation [217]:

$$E_{\mathrm{Rn}} = \bar{C}_{\mathrm{Rn}} \times F_e \times T_{\mathrm{a}} \times D_{\mathrm{Rn}} \qquad (3.7)$$

Where E_{Rn} is the annual effective dose of ^{222}Rn (Sv y^{-1}), T_{a} is the annual work time = 7000 h y^{-1}, D_{Rn} the dose conversion factor for ^{222}Rn decay products [9 nSv h^{-1} (Bq m^{-3})$^{-1}$] and $F_e$is the equilibrium factor. The value of F_e was assumed to be 0.4 [217].

CR-39 detectors have a good capability to register tracks at different levels of registration sensitivity. Therefore, we used them to measure the effective radium content for

the same samples [190], [206], [218]. After the sealing of the can, the activity concentration of radon begins to increase with time *T* according to the following equation [206], [219]:

$$C_{\mathrm{Rn}} = C_{\mathrm{Ra}}\left(1 - e^{-\lambda_{\mathrm{Rn}}\ T}\right) \qquad (3.8)$$

Where $C_{\mathrm{Ra}}$is the effective radium content [190], [193], [206], [209] of the building material sample. Also C_{Ra} is called the effective radium concentration [220], [221], as this is the fraction of total radium which contributes to radon exhalation [222]. The effective radium content was calculated by using the following relation [205], [223]:

$$C_{Ra}(\mathrm{Bq\ kg^{-1}}) = \frac{h\ \rho\ A}{K T_e\ M} \qquad (3.9)$$

Where *h* is the distance between the detector and the top of the solid sample (Varies from one sample to another), ρ is the radon track density (track $\mathrm{cm^{-2}}$) and T_e is the effective exposure time (in days), which is related to the actual exposure time *T* and decay constant for^{222}Rn according to the following [224], [225]:

$$T_e = \left[T - \frac{1}{\lambda_{\mathrm{Rn}}}\left(1 - e^{-\lambda_{\mathrm{Rn}}\ T}\right)\right] \qquad (3.10)$$

Radon exhalation from the building materials contributes towards indoor radon. In this regard, previous studies have been carried out in different parts of the world and extensive data are available in the literature. In the current survey, most of the houses in the studied area are built of blocks (mixtures of cement sand and gravel aggregates in definite proportions). Some of the houses were made up of bricks and Sand and most of them also contain interior decoration made of ceramics, gypsum, marble, and granite. These houses are in fact constructed on the Earth's crust, which is essentially composed of rocks and soil.

3.8 Calculations of Radon in Soil and Fertilizers

3.8.1 Radon in Soil Air

Radon concentration in soil air was calculated by using the relation (3.3).

3.8.2 Radon in Fertilizers

Radon exhalation rate, concentration, the effective radium content for fertilizers was calculated by using the same relations (3.3), (3.4), (3.5) and (3.9).

In section 3.7 and 3.8, measurements have been made by using the parameters in the Table 3-1.

Table 3-1: Some parameters used for measurement: radon concentrations, exhalation rate, annual effective doses and effective radium content.

Parameters / Experimental	K Calibration factor (Track. $cm^{-2}.d^{-1}$/ $Bq.m^{-3}$)	A Area of the field (cm^2)	*T* The exposure time (Months)
Building materials	0.16 (calibrated in previous work [226]–[228])	1.886×10^{-3}	3
Soil	0.192 (calibrated in previous work [229], [230])	1.598×10^{-3}	1
Fertilizers	0.16 (calibrated in previous work [226]–[228])	1.886×10^{-3}	3

CHAPTER 4: RESULTS AND CONCLUSION

4.1. Building Materials

4.1.1 Building Materials in Yemen

The surface radon exhalation rates from the solid slab samples ranged from 275.24 ± 5.43 to 1267.6 ± 11.84 mBq $m^{-2}h^{-1}$, with a mean of 661.69 ± 8.56 mBq$m^{-2}h^{-1}$, whereas the surface exhalation rates from the powdery samples ranged from 178.90 ± 6.76 to 1079.53 ± 9.70 mBq $m^{-2}h^{-1}$, with a mean of 462.46 ± 10.02 mBq $m^{-2}h^{-1}$ (see Table 4-1 and 4-2). The mass radon exhalation rates from the powdery samples ranged from 5.51 ± 0.2 to 33.25 ± 0.30 mBq $kg^{-1}h^{-1}$, with a mean of 4.24 ± 0.31mBq $kg^{-1}h^{-1}$ (see Table 4-2). The values of exhalation rates of building materials vary from one building material to another. This variation can be attributed to the difference in the nature of the samples, and the effective radium content in the samples.

To estimate the contribution of building materials to indoor radon concentrations [206], a room with an air exchange rate of 0.5 h^{-1} was assumed as mentioned above. The radon concentration contribution to indoor radon in the room due to exhalation from the solid slab samples building materials was found to range from 1.10 ± 0.02 to 5.07 ± 0.05 Bq m^{-3}, with a mean of 2.65 ± 0.03Bq m^{-3} (see Table 4-1). The smallest accumulated radon concentration per day in a room with its entire floor and walls decorated with M2, denoting Yemen marble, would be 26.42 Bq m^{-3}. Thus, on average, the sample M2 would contribute only slightly to the radon concentration in Yemen homes. On the other hand, the largest accumulated radon concentration per day was 121.656 Bq m^{-3} for sample Gra1, denoting Yemen granite.

Table 4-1: Radon contribution to indoor radon, surface exhalation rate, and annual effective doses, together with the statistical uncertainties (1σ) for the solid slab differentbuilding material samples.

S . No	Sample ID	Radon surface exhalation rate ($mBq\ m^{-2}h^{-1}$)	Radon contribution ($Bq\ m^{-3}$)	Annual effective dose ($\mu Sv\ y^{-1}$)
1	M1	527.53±10.84	2.11±0.04	53.18
2	M2	275.24±5.43	1.10±0.02	27.74
3	M3	513.77±6.87	2.06±0.03	51.79
4	M4	336.40±6.28	1.35±0.03	33.91
5	Gra1	1267.61±11.84	5.07±0.05	127.78
6	Gra2	1079.53±13.51	4.32±0.05	108.82
7	Ce1	827.23±11.63	3.31±0.05	83.39
8	Ce2	564.23±6.46	2.26±0.03	56.87
9	Ce3	382.27±8.15	1.53±0.03	38.53
10	Ce4	342.52±5.70	1.37±0.02	34.53
11	RB1	519.89±8.00	2.08±0.03	52.40
12	RB2	593.28±6.04	2.37±0.02	59.80
13	T	758.43±6.87	3.03±0.03	76.45
14	CB1	889.93±8.27	3.56±0.03	89.70
15	CB2	1047.42±12.53	4.19±0.05	105.58
M: marble, RB: red brick, CB: concrete block, Gra: granite, Ce: ceramic, T: tiles				

For the powdery materials, they ranged from 0.72 ± 0.03 to $4.32 \pm 0.04\ Bq\ m^{-3}$. The sand sample (Sa1) showed the lowest value, and the daily accumulated radon concentration in a room was estimated at $17.16\ Bq\ m^{-3}$. On the other hand, the soil sample (So2) gave the highest value observed in this type of study. The daily accumulated radon concentration in a room was estimated to be $103.63\ Bq\ m^{-3}$.

Table 4-2: Radon contribution to indoor radon, surface and mass exhalation rates, annual effective doses and effective radium content, together with the statistical uncertainties (1σ) for the powdery different building material samples.

S. No	Sample ID	Size (cm^3)	Radon surface exhalation rate ($mBq\ m^{-2}h^{-1}$)	Radon contribution ($Bq\ m^{-3}$)	Radon mass exhalation rate ($mBq\ kg^{-1}h^{-1}$)	Effective radium content ($Bq\ kg^{-1}$)	Annual effective dose ($\mu Sv\ y^{-1}$)
1	C1	342.65	207.96±7.88	0.83±0.03	6.40±0.24	0.69	20.96
2	C2	338.80	243.12±6.07	0.97±0.02	7.49±0.19	0.79	24.51
3	C3	338.80	507.66±10.73	2.03±0.04	15.64±0.33	1.66	51.17
4	WC1	338.80	276.76±6.07	1.11±0.02	8.52±0.19	0.90	27.90
5	WC2	342.65	455.67±10.85	1.82±0.04	14.03±0.33	1.50	45.93
6	WC3	342.65	553.53±9.39	2.21±0.04	17.05±0.29	1.83	55.80
7	Sa1	346.50	178.90±6.76	0.72±0.03	5.51±0.21	0.60	18.03
8	Sa2	346.50	428.14±10.41	1.71±0.04	13.19±0.32	1.43	43.16
9	Sa3	346.50	339.46±6.62	1.36±0.03	10.45±0.20	1.13	34.22
10	Sa4	346.50	633.04±35.51	2.53±0.14	19.50±1.09	2.11	63.81
11	Sa5	346.50	463.31±6.76	1.85±0.03	14.27±0.21	1.55	46.70
12	Gy1	331.10	325.70±6.76	1.30±0.03	10.03±0.21	1.04	32.83
13	Gy2	327.25	441.91±10.98	1.77±0.04	13.61±0.34	1.39	44.54
14	Gr1	327.25	526.01±7.22	2.10±0.03	16.20±0.22	1.66	53.02
15	Gr2	346.50	449.55±8.62	1.80±0.03	13.85±0.27	1.50	45.31
16	Gr3	342.65	474.02±10.52	1.90±0.04	14.60±0.32	1.57	47.78
17	So1	342.65	740.08±9.57	2.96±0.04	22.79±0.29	2.44	74.60
18	So2	342.65	1079.53±9.70	4.32±0.04	33.25±0.30	3.56	108.82

C: cement, WC: white cement, Sa: sand, Gy: gypsum, Gr: gravel, So: Soil

To study the contribution of building materials to indoor radon concentrations with air changes per hour (ACH), two scenarios were considered:

Case I: Radon Exhalation from a Granite Countertop

Consider a granite countertop (0.67 m×2.50 m× 0.025 m) installed in a kitchen with an area of 20 m^2 and a height of 2.5 m [231], with the assumption that 25% of the kitchen volume is occupied by kitchen wares and furniture, and assuming as well that the kitchen is ventilated with the minimum required ACH of 0.3 h^{-1}, for granite countertops having the highest radon exhalation rate (30.42 $Bq\ m^{-2}d^{-1}$) observed in this study, the radon concentration in the kitchen will be 0.37 $Bq\ m^{-3}$. It can be concluded that kitchen granite countertops, under normal ventilation, contribute very little to the radon concentration for the range of radon exhalation rates reported here. Similar assessments were reported in the literature [198], [231]. Based on more data available recently, it was assessed that the average radon concentration in Yemen homes is 42 $Bq\ m^{-3}$ [232]. It indicates that on average, granite countertops could contribute to less than 1% of radon concentration in Yemen homes.

Case II: Radon Exhalation from a floor area

If a room has a floor area of 20 m^2 and a height of 2.5 m as assumed in scenario 1 and its entire floor is decorated with ceramic tiles, slate or granite, and 10% of the room volume is occupied by furniture, the radon concentration, Bq m^{-3}, due to radon exhalation from the floor can be determined by Eq.(3.6). The resulting radon concentrations are calculated for materials of various radon exhalation rates and for various air change rates, as shown in Table 4-3. One can infer from Table4-3 that even if the entire floor was covered with a material of a relatively high radon exhalation rate, as granites studied here 30.42 Bq $m^{-2}d^{-1}$, it would contribute only 1.83 Bq m^{-3} to a tightly sealed house (minimum required ACH of 0.3 per hour). Higher ventilation rates will reduce the contribution proportionally. In the worst case scenario and when the mechanical ventilation system failed completely and no natural ventilation alternative was provided (ACH = 0), the indoor radon concentration would rise is higher than the world average value of 40 Bq m^{-3} as reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR2000) [9][233] but not above an action level of 300 Bq m^{-3} for radon in dwellings ICRP 2014[138] or even the one adopted by some countries, i.e. 200 Bq m^{-3} [234].

Table 4-3: Estimate steady-state radon concentration (Bqm^{-3}) due to radon exhalation fromfloor material with various air change rates.

Sample	E_A (Bq $m^{-2}d^{-1}$)	ACH = 3	ACH = 1	ACH = 0.3	ACH = 0.15	ACH = 0
Marble	13.02	0.08	0.24	0.78	1.53	31.97
Ceramic	19.85	0.12	0.36	1.20	2.33	48.75
Tiles	21.92	0.13	0.40	1.32	2.58	53.81
Granite	30.42	0.19	0.56	1.83	3.58	74.69

The effective radium content (ERC) in powdery samples was calculated using Eq. (9). And the results are provided in Table 4-2. ERC range from 0.6 to 3.56 Bq $kg^{-1}$with a mean value of 1.52Bq kg^{-1}.All the values of radium content in all samples under test were found to be quite lower than the permissible value of 370 Bq kg^{-1} recommended by Organization for Economic Cooperation and Development (OECD) [235][236].

Finally, the assessment of the annual effective dose expected to be received by populations due to radon and its progenies were based on the calculations of radon exhalation rates. The minimum, maximum and mean annual effective doses were found to be 27.74 μSv y^{-1}, 127.78 μSv y^{-1}, and 66.70 μSv y^{-1} for the slabs samples, and 18.03 μSv y^{-1}, 108.82 μSv y^{-1} and 46.62 μSv $y^{-1}$for the powdery samples. The slabs samples giving the

lowest values of the annual effective dose were marble (M2) from Yemen (Taiz); the lowest dose value from the powdery samples was attributed to the Sa1, denoting Yemen sand. The highest dose from the slabs samples came from granite in Yemen and the highest dose in the powdery samples came from So2, denoting Yemen. The results obtained are shown in Tables 4-1 and 4-2. The results of the current study have been compared with already published data (Table 4-8).

The annual effective dose in this study for all samples varies from 18.03 to 127.77 $\mu Sv\ y^{-1}$ with an average value of 55.74 $\mu Sv\ y^{-1}$, which is lower than the action level of 3–10 $mSv\ y^{-1}$ recommended by International Commission on Radiological Protection (ICRP, 1993b,2014) [237][138]. Also it is lower than the global average of 1.15 $mSv\ y^{-1}$ as estimated by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2008)[238][233].

4.1.1.1 CONCLUSION

- The two highest values of radon exhalation observed in the studied solid slabs samples came from the Yemeni granite and the Chinese granite. For this reason, these two materials are not favored for decorative use and should be replaced by other alternatives.

  For the powdery sample, the soil was found to have a relatively high level of radon exhalation. However, there exists only a small number of houses which are made of mud alone. Without ventilation, elevated radon levels could arise. In general, All the results for radon exhalation are less than the world average value of 57.600 $Bq\ m^{-2}\ h^{-1}$, recommended by the (UNSCEAR, 2000) [239], [240].

  The contribution of the building materials to the indoor ^{222}Rn concentration is lower than the value recommended by ICRP [2014] for the investigation level of 300 $Bq\ m^{-3}$ or even the one adopted by some countries, i.e. 200 $Bq\ m^{-3}$ [234] .
- The effective radium content and annual effective dose equivalents are within the safe limits recommended by (OECD) [235] and ICRP,1993 b,2014) [138][237], respectively.

  Generally speaking, building materials used in home construction and decoration make no significant contribution to indoor radon for a house with adequate air exchange. It is possible to establish the data base for all building materials sold in the

market by using this technique instead of spending much money and resources to perform a large-scale nationwide indoor radon screening measurement.

4.1.2 Building Materials in Morocco

In the current survey, the surface exhalation rate, the mass exhalation rate and the annual effective dose for the building materials were given by Table 4-4 and 4-5.The surface radon exhalation rates from the solid slab samples ranged from 37.92 ± 1.87 mB q m^{-2} h^{-1}to1545.60 ± 21.26 mB q m^{-2} h^{-1}, with a mean of 232.68 ± 4.25 mB q m^{-2} h^{-1}, where the highest value for granite (Gra1), is roughly equal to that value obtained in a previous study [241]–[243], whereas the surface exhalation rates from the powdery samples ranged from 99.08 ± 5.06to 1400.64 ± 31.26 mB q m^{-2} h^{-1}, with a mean of 317.40 ± 4.41mB q m^{-2} h^{-1}.The max value, denoting cement Portland 45 (C5). This value is less than the value obtained by a previous study[218](see Table 4-8). Cement is one of the most commonly used building materials for construction. Calcium carbonate (lime), silica, alumina, iron oxide and fly ash are the raw materials for the cement (Portland). Hence, the activity concentrations of radon in cement samples depend on the activity of constituents. However, addition of fly ash in cement also leads to high radon activity [244] and the least value, denoting gravel (Gr2).

The mass of the radon exhalation rates from the powdery samples ranged from 3.05 ± 0.16 to 43.14 ± 0.96 mBq kg^{-1} h^{-1} with a mean of 10.23 ± 0.77 mBq kg^{-1} h^{-1} , where the highest value is cement Portland 45(C5). The average value of the mass exhalation rates (E_M) in this study is remarkably lower than those reported for India [245] and Libya [206]; whereas this value of mass exhalation rates (E_M) is higher than those reported for Lebanon [246] and Sudan [247] (see Table 4-8).

Table 4-8 shows the comparison between surface exhalation rates (E_A) and mass exhalation rates (E_M) in the present study with those obtained elsewhere. Based on the comparison, it has been observed that there is also a variation in the values of radon levels among countries. This variation is due to the difference in the effective radium content, uranium concentration[247] and the porosity of various materials [248], [249].

To estimate the contribution of building materials to indoor radon concentrations, a room with an air exchange rate of 0.5 h^{-1} was assumed as mentioned above. The radon concentration contribution to indoor radon in the room due to the exhalation from the solid slab of building material samples ranged from 0.15 ± 0.03 to 6.18 ± 0.11 Bq m^{-3}, with a mean of 0.93 ± 0.27 Bq m^{-3}. The smallest accumulated radon concentration per day in a room with

its entire floor and walls decorated with Ce1, denoting Moroccan ceramics, would be 3.64 Bq m^{-3}. Thus, on average, the sample Ce1 would contribute only slightly to the radon concentration in Moroccan homes. On the other hand, the largest accumulated radon concentration per day was148.41 Bq m^{-3} for samples Gra1, denoting Italian granite. For the powdery materials they ranged from 0.40±0.05 to 5.60±0.13 Bq m^{-3}. The gravel sample (Gr2) showed the lowest value, and the daily accumulated radon concentration in a room was estimated to be 9.51 Bq m^{-3}. On the other hand, the cement Portland 45(C5) gave the highest value observed in this type of study. The daily accumulated radon concentration in a room was estimated at 134.46 Bq m^{-3}.

Table 4-4: Radon contribution to indoor radon, surface exhalation rates and annual effective doses for the slab different building material sample.

S. No	Sample ID	Radon surface exhalation rate (mB q $m^{-2} h^{-1}$)	Radon contribution (Bq m^{-3})	Annual effective dose (μSv y^{-1})
1	RB	136.39+5.54	0.55+0.06	13.75
2	CB	72.17+3.78	0.29+0.05	7.28
3	Ce1	37.92+1.87	0.15+0.03	3.82
4	Ce2	129.67+3.86	0.52+0.05	13.07
5	Ce3	152.30+6.85	0.61+0.06	15.35
6	Ce4	102.75+5.15	0.41+0.05	10.36
7	Ce5	115.60+3.84	0.46+0.05	11.65
8	Ce6	75.23+3.05	0.30+0.04	7.58
9	Ce7	116.82+5.29	0.47+0.05	11.78
10	T	85.02+4.31	0.34+0.05	8.57
11	M1	64.22+4.21	0.26+0.05	6.47
12	M2	78.29+4.80	0.31+0.05	7.89
13	M3	122.94+5.13	0.49+0.05	12.39
14	M4	51.99+3.06	0.21+0.04	5.22
15	Gra1	1545.60+21.26	6.18+0.11	155.83
16	Gra2	516.22+14.27	2.05+0.09	51.71
17	Gra3	263.61+12.38	1.05+0.08	26.55
18	Gra4	343.74+10.76	1.37+0.08	34.54
19	Gra5	410.41+10.60	1.64+0.08	41.39
M: marble, RB: red brick, CB: concrete block, Gra: granite, Ce: ceramic, T: tiles				

Table 4-5: Radon contribution to indoor radon, surface and mass exhalation rates, annual effective doses, effective radium content for the powdery different building material sample.

S. No	Sample ID	Radon surface exhalation rate (mB q m^{-2} h^{-1})	Radon contribution (Bq m^{-3})	Radon mass exhalation rate (mBq kg^{-1} h^{-1})	Effective radium content (Bq kg^{-1})	Annual effective dose (μSv y^{-1})
1	C1	467.29+12.20	1.87+0.08	14.392501	1.53	47.10
2	C2	442.82+15.01	1.77+0.09	13.638967	1.45	44.64
3	C3	353.52+10.95	1.41+0.08	10.888567	1.15	35.64
4	C4	315.60+10.02	1.26+0.08	9.720590	1.03	31.81
5	C5	1400.64+31.26	5.60+0.13	43.139826	4.57	141.18
6	WC1	222.63+6.77	0.89+0.06	8.571450	0.91	22.44
7	WC2	310.71+8.67	1.24+0.07	11.962354	1.24	31.32
8	GY1	166.98+6.25	0.67+0.06	6.428588	0.68	16.83
9	GY2	130.28+5.51	0.52+0.06	5.015711	0.53	13.13
10	GY3	122.33+4.00	0.49+0.05	4.709588	0.50	12.33
11	Sa1	264.23+7.25	1.06+0.06	8.138168	0.86	26.63
12	Sa2	204.29+7.12	0.82+0.06	6.292010	0.67	20.59
13	Gr1	108.87+3.72	0.44+0.05	3.353227	0.36	10.97
14	Gr2	99.08+5.06	0.40+0.05	3.051813	0.32	9.99
15	So1	162.69+4.82	0.65+0.05	5.011002	0.53	16.40
16	So2	306.43+7.07	1.23+0.06	9.438014	1.00	30.89
C: cement, WC: white cement, Sa: sand, GY: gypsum, Gr: gravel, So: Soil						

To assess the contribution of building materials to indoor radon concentrations, two scenarios were considered. In all cases, a tightly sealed house with an air change rate of a given ACH was assumed:

Case I: Radon Exhalation from a Floor Area

If a room has a floor area of 20 m^2 and a height of 2.5 m its entire floor decorated with ceramic tiles or slate or granite and 10% of the room volume is occupied by furniture, the radon concentration, Bq m^{-3}, due to radon exhalation from the floor can be determined by Eq. (3.6). The resulting radon concentrations are calculated for materials of various radon exhalation rates and for various air change rates, as shown in Table 4-6.

One can see from Table 4-6 that even if the entire floor was covered with a material of a relatively high radon exhalation rate, like granites studied here 37.09 Bq $m^{-2}d^{-1}$, it would contribute only 2.234 Bq m^{-3} to a tightly sealed house (minimum required ACH of 0.3 per hour)[231]. Higher ventilation rates will reduce the contribution proportionally. In the worst case when the mechanical ventilation system failed completely and no natural ventilation alternative was provided (ACH = 0), the indoor radon concentration would rise is higher than the world average value of 40 Bq m^{-3} as reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR2000) [9][233] but not above an action level

of 300Bq m^{-3} for radon in dwellings ICRP 2014 [138] or even the one adopted by some countries, i.e. 200 Bq m^{-3} [234]

Table 4-6 : Estimate steady-state radon concentration (Bqm^{-3}) due to radon exhalation fromfloor material with various air change rates.

Sample	E_A (Bq $m^{-2}d^{-1}$)	ACH = 3	ACH = 1	ACH = 0.3	ACH = 0
M	2.95	0.018	0.054	0.178	7.245
Ce	3.66	0.023	0.067	0.22	8.975
Gra	37.09	0.228	0.682	2.234	91.09

Case II – Radon Exhalation from a Building Material Stockpile

Let us consider 300 slabs of building material (1.5 m wide, 3.0 m long and 2.5 cm thick) stored in a space having a floor area of 100 m^2 and 3.5 m from floor to ceiling. The 300 slabs have a total surface of 2700 m^2, this total surface is the effective exhalation surface, i.e. slabs are stacked with air gaps that do not inhibit emission. The volume is about 316.4 m^3 of the room volume. The radon concentration, Bq m^{-3}, due to exhalation from those stockpiled materials, can be determined by Eq. (3.6) [231].

For the case examined here, radon exhaled from 300 slabs does not constitute any serious problem where the slabs of various types were stored and the average radon concentration was less than the world average value of 40 Bq m^{-3} as reported by the UNSCEAR2000 [9] [226][250]. See Table 4-7.

Table 4-7: Estimated steady-state radon concentration (Bq m^{-3}) due to radon exhalation from 300 slabs of building materials (1.5 m × 3 m × 0.025 m) stored in a room (350 m^3) with various air exchange rates of ACH.

Sample	E_A (Bq $m^{-2}d^{-1}$)	ACH =10	ACH =5	ACH =2	ACH=1
M	2.95	0.10	0.21	0.52	1.04
Ce	3.66	0.13	0.26	0.65	1.29
Gra	37.09	1.32	2.63	6.57	13.09

In general, the resulting radon concentration, $\bar{C}_{\mathrm{Rn}}$, is proportional to the radon exhalation rate, E, and the total area, A of stockpiled materials. For the given E and A, $\bar{C}_{\mathrm{Rn}}$ increases with decreasing air volume, V, of a room i.e. with increased percentage of the space occupied with such slabs [231].

The effective radium content (ERC) in powdery samples was calculated using Eq. (3.9). As shown in Table 4-5, the ERC ranged from 0.32 to 4.57 Bq kg^{-1} with a mean value of

1.1 Bq kg^{-1}. Overall, the values of radium concentration in building materials samples in the present study are much lower than the permissible value recommended by the OECD [235].

In order to estimate the annual effective doses in indoor air, the contribution of radon from building materials to indoor radon is determined by using the measured area radon exhalation rates.

The calculated values of the annual effective dose equivalents vary from 3.82 to 155.83 μSv y^{-1} with an average value of 27.35 μSv y^{-1}, which is lower than the action level of 3–10 mSv y^{-1} recommended by (ICRP,1993,2014) [138][237], also it is lower than the global average of 1.15 mSv y^{-1} as estimated by (UNSCEAR 2008) [238][233].

4.1.2.1 Conclusion

Results obtained from the current study show that:

For the powdery sample and the slab sample, the cement (C5) and the granite (Gra1), all have high level of radon exhalation. Without ventilation, elevated radon levels could arise. In general, the current results are within the world-wide range of values found in building materials, and this range is within the safe limits proposed in the report issued by UNSCEAR (2000) [239].

It is recommended that the radon exhalation rate should be measured for all building materials and a standard code should be placed on all products. This will minimize the indoor radon concentration in new construction, especially in new locations.

The values of effective radium content are less than the permissible value of 370 Bq kg^{-1} recommended by Organization for Economic Cooperation and Development [235].

The annual effective doses is, on average, below the action level recommended by(ICRP,1993 b,2014) [138][237] and (UNSCEAR 2008) [238][233].

Generally speaking, building materials used in home construction and decoration make no significant contribution to indoor radon for a house with adequate air exchange.

Table 4-8: Comparison between surface exhalation rates (E_A)and mass exhalation rates (E_M) of some building materials in different countries.

Material	countries	E_A (Range) (mB q m^{-2} h^{-1})	E_M (Range) (mBq kg^{-1} h^{-1})	Reference
Granite	USA	160 – 1480		[243]
	KSA	1500		[242]
	Libya	733.2 – 17221.2		[206]
	Greece	1240 – 3540		[118]
	Hungary	1100		[241]
	KSA	333 – 1250		[251]
	Morocco	263.61– 1545.60		Present work
	Yemen	1079.53−1267.61		Present work
Ceramic	Sudan	240 – 949		[247]
	Canada	8.33 – 125		[231]
	Libya	69.5 – 73.7		[206]
	Morocco	37.92 – 152.30		Present work
	Yemen	382.27 − 827.23		Present work
Marble	Algeria	35 – 66		[252]
	Pakistan	120		[102]
	Italy	65.4		[203]
	Canada	4.17 – 25		[231]
	Libya	142.3 − 970.6		[206]
	Morocco	51.99 – 122.94		Present work
	Yemen	275.24 − 527.53		Present work
Gravel	Canada	45.83 – 4245.83		[198]
	Pakistan	168 – 322		[253]
	Lebanon	44 – 2226	0.4 – 20.0	[246]
	Morocco	99.08 – 108.87	3.05 – 3.35	Present work
	Yemen	449.55 − 526.01	13.85 − 16.20	Present work
Sand	Pakistan	3900 – 17000		[254]
	Pakistan	292		[102]
	Pakistan	205 – 291		[255]
	Pakistan	366 – 649		[253]
	Libya	722.2	27.4	[206]
	Sudan	391	4.64	[247]
	Lebanon	512	6.7	[246]
	India	423.2 – 1636.3		[244]
	Morocco	204.29 – 264.23	6.29 – 8.14	Present work
	Yemen	178.90 − 633.04	5.510 − 19.497	Present work
Gypsum	Egypt	26.28		[189]
	Libya	147.5 – 365.5	17.4 – 43.1	[206]
	Lebanon	65	0.86	[246]
	Morocco	122.33 – 166.98	4.71 – 6.43	Present work
	Yemen	325.70 − 441.91	10.031 − 13.610	Present work
Soil	Pakistan	2200 – 2800		[254]
	Syria	72 000 – 32 400 000		[256]
	Moroccan	3 – 145		[89]
	India	246.63 – 1100		[257]
	Libyan	58.3 – 506.9	1.2 – 17.4	[202]
	KSA	458 – 8400	135 – 251	[225]
	India		6 – 56	[195]
	Pakistan	114 – 416		[255]
	Egypt	48 0−15370	8.31 − 233.70	[190]

Material	countries	E_A (Range) (mB q m^{-2} h^{-1})	E_M (Range) (mBq kg^{-1} h^{-1})	Reference
	Morocco	162.6 – 306.43	5.01 – 9.44	Present work
	Yemen	740.08 −1079.53	22.794 − 33.248	Present work
Cement	Austria	270 – 660		[258]
	Lebanon	197	2.6	[246]
	Egypt	2549 – 7557		[218]
	Sudan	379	4.5	[247]
	India	263.3 – 401.6	12.1 – 18.4	[245]
	Libya	523.2	20.4	[206]
	Morocco	315.60 – 1400.64	9.72 – 43.14	Present work
	Yemen	207.96−507.66	6.405 − 15.64	Present work
White cement	Libya	550.9	20.8	[206]
	India	112.40	3.40	[259]
	India	417 – 437	13.8 – 20.1	[245]
	Lebanon	35	0.46	[246]
	Morocco	222.63–310.71	8.57–11.96	Present work
	Yemen	276.76 − 553.53	8.524 − 17.048	Present work
Brick	Pakistan	184 - 231		[253]
	Pakistan	245 - 365		[255]
	Libya	123.7 − 1625.1		[206]
	Yemen	519.89 − 593.28		Present work

4.1.3 General Discussion for Building Materials

We found that the highest value annual effective dose in the powdery samples of Yemeni soil is (91.71μSv y^{-1}) whereas it in the Moroccan soil (23.645μSv y^{-1}) (see Fig. 4-1). This is due to the fact that the Yemeni soil sample has been taken from an agricultural area which is exposed repeatedly to different fertilizers that may contain quantities of radioactive elements. Many studies have shown that there is a direct relationship between the increasing amount of radon in the soil and the fertilizers used [260] [24]. The soil studied in Morocco belong to the city of Rabat and taken from a depth of 30-40 cm and, at this distance the soil could have been exposed to transportation from one place to another at the time of building and thus the samples taken from those areas does not represent the city of Rabat soil. Also, the soil of the city of Rabat is not agricultural, so fertilizers are not added to it from time to time. This reason makes us interested more in studying the radon concentration in Yemen soil and the fertilizers used to help crops grow.

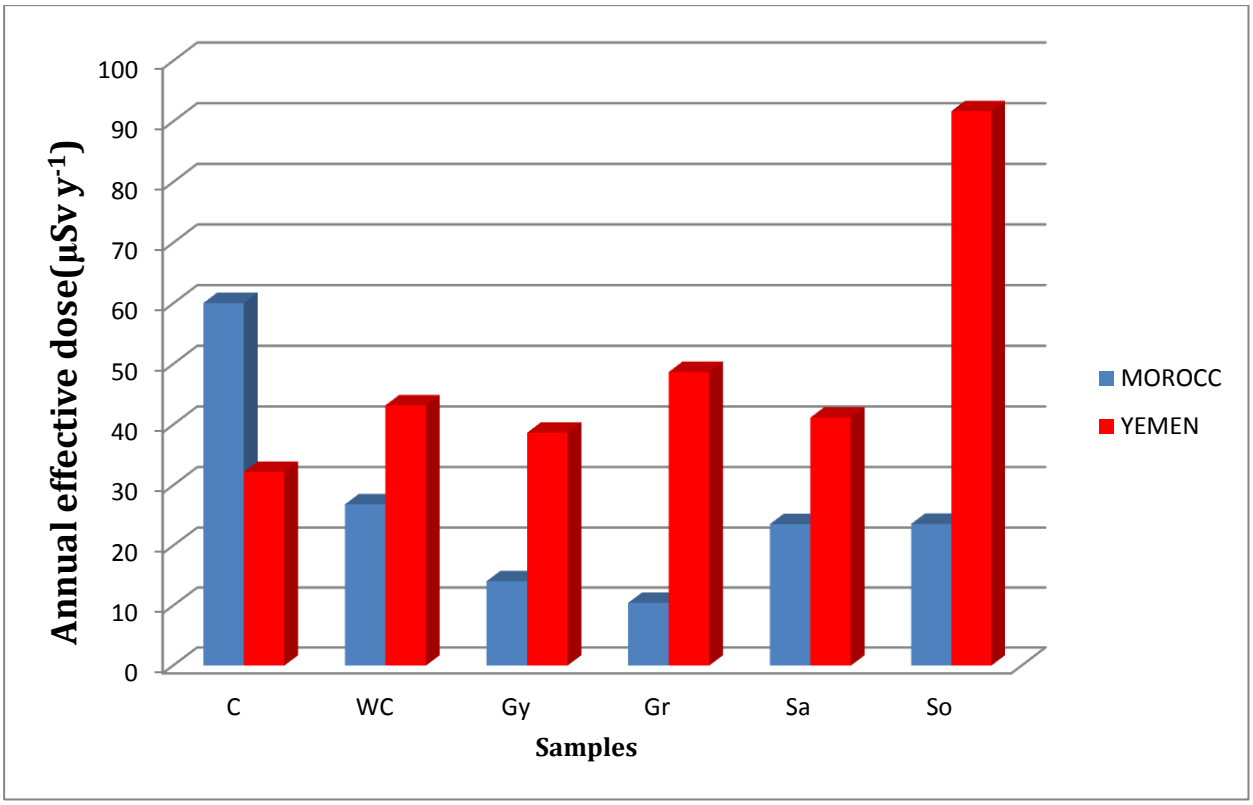

Fig.4-1: Comparison between the annual effective dose of building material (powdery) samples in Yemen and Morocco

It is very clear from the Fig.4-1 that the annual effective dose rate from the Moroccan cement is high and this is due to one types of cement (C5) which contains a large proportion of radon concentration as shown in the Table 4-5.

The highest annual effective dose has been found from slab samples in Yemen or Morocco were in granite and this is something expected. It has been found in many studies that granite contains a high concentration of radon [261][262].

The second highest value found was in the Yemeni concrete block (CB), See Fig.4-2. A concrete block is a mixture of cement, sand and gravel. Therefore, an increase in the annual effective dose of cement, gravel, and sand together, or an increase in the annual effective dose for one of them (such as gravel in this study) leads to an increase in the total annual effective dose of concrete block.

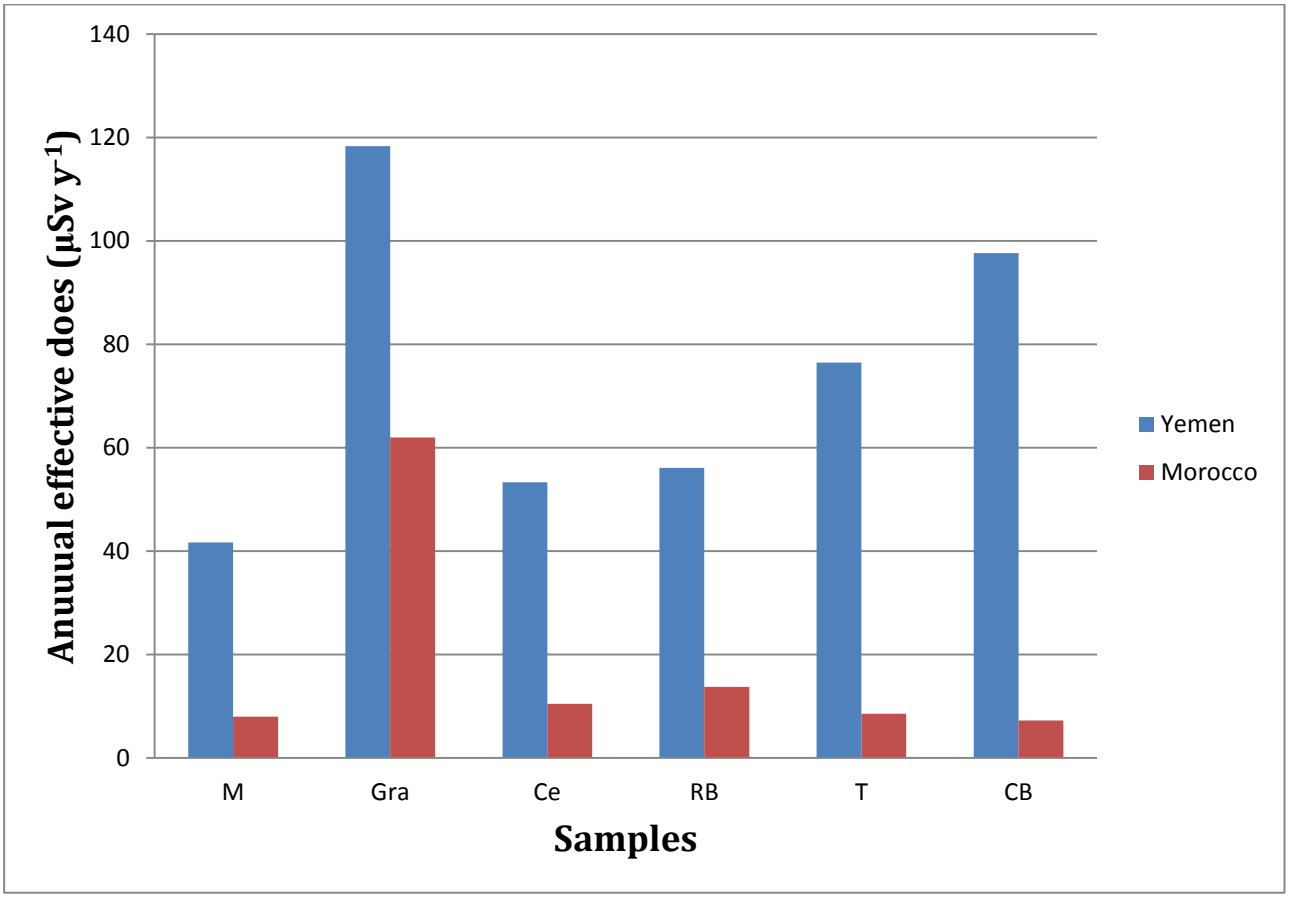

Fig.4-2: Comparison between the annual effective dose of building material (slab) samples in Yemen and Morocco.

Fig. 4-2 shows that the tile samples have a high dose value and this may be due to their components. However, it is just one sample and it cannot be generalized to the rest of the tiles samples present in the market. Some samples are few in number; this is due to the damage or loss of some of detectors for samples. This can be attributed to the duration of the work and the many stages of the experiment involved.

Generally, it is noted from Fig. 4-1 and Fig. 4-2 that the annual effective dose from building materials samples (powder and slab) in Yemen are higher than the doses from the building materials in Morocco. This might also be due to the location and the nature of the study area. Ibb province is an agricultural and mountainous region [263][264]. The type of rocks in this region may have a significant impact on the results.

Also, the quality of the building materials imported to Yemen or Morocco varies according to the source country and the quality standards in each country.

4.1.4 General conclusions and recommendations

From our study, the effective dose given by all investigated building material samples was found to fall within the range of the recommended action level (3–10) $mSv.y^{-1}$ as given by (ICRP,1993 b,2014), However:

- One of the Moroccan cement types was found to have a relatively high dose of radiation. It is therefore important for buildings whose walls mainly consist of cement to take extra care through ensuring continuous ventilation and painting the walls with materials that prevent radon from escaping into the house to avoid exposure to radon.

- Soil samples in the study area of Yemen have high value of radiation dose and this poses a serious threat to the population in the future, especially when fertilizers are added periodically.

- In both studies, granite has a high value of radiation dose. It is therefore important to limit or reduce the use of granite, especially in the closed spaces.

In order to protect the population, we recommend that Yemen and Morocco should join the countries that impose standards and guideline on mitigation or prevention of radon risks or set standards for radon exposure.

Those countries include Austria, Belgium, China, the Czech Republic, Finland, France, Ireland, Latvia, Norway, Russia, Sweden, Switzerland, the United Kingdom, and the United States of America [265].

When developing guidelines and standards for Radon prevention and mitigation, it is important to consult radon mitigation contractors, building researchers as well as other building and construction professionals.

Flater and Spencer (1994) have shown that if these guidelines and standards become part of building codes, inspection procedures are needed to insure compliance [152].

Lung cancer exists at levels of long-term average radon concentration below 200 Bq m^{-3} (ICRP, 2010).

As a consequence, the aim should be to reduce both the overall risk for the population and, for the sake of equity, the highest individual exposures to levels that are as low as reasonably achievable.

However, the total elimination of radon exposure is not feasible.

4.2. Concentration of Radon Gas in the Soil

4.2.1 Radon Concentration at Horizontal Study (50 cm)

For the horizontal study (location A, location B), the minimum radon concentration is 1.59 KBq m^{-3}, the maximum radon concentration is 7.10 KBq m^{-3}, and the average radon concentration is 3.887 KBq m^{-3} (See Table 4-9).

The positional changing of radon concentration in soil at a particular depth, as it has been found in this study (see Fig.4-3), is attributed to many factors that determine the radon emission from soil. One of the most important factors is that radon is distinguished by its short diffusion distance. Therefore, radon exists close to the place that it is emitted from. Other factors include the variations of the concentration of gases, which transport radon, with changing position like (co_2), variations of distribution of fertilizers in soil and variations to cultivate. Perhaps there are other factors that are related to position, such as the existence rate of uranium and radium in soil or in bedrock, weathering, porosity, permeability and moisture content of soil. Therefore, the inconstant positional change of radon concentration in soil horizontally results from the existence of these factors together.

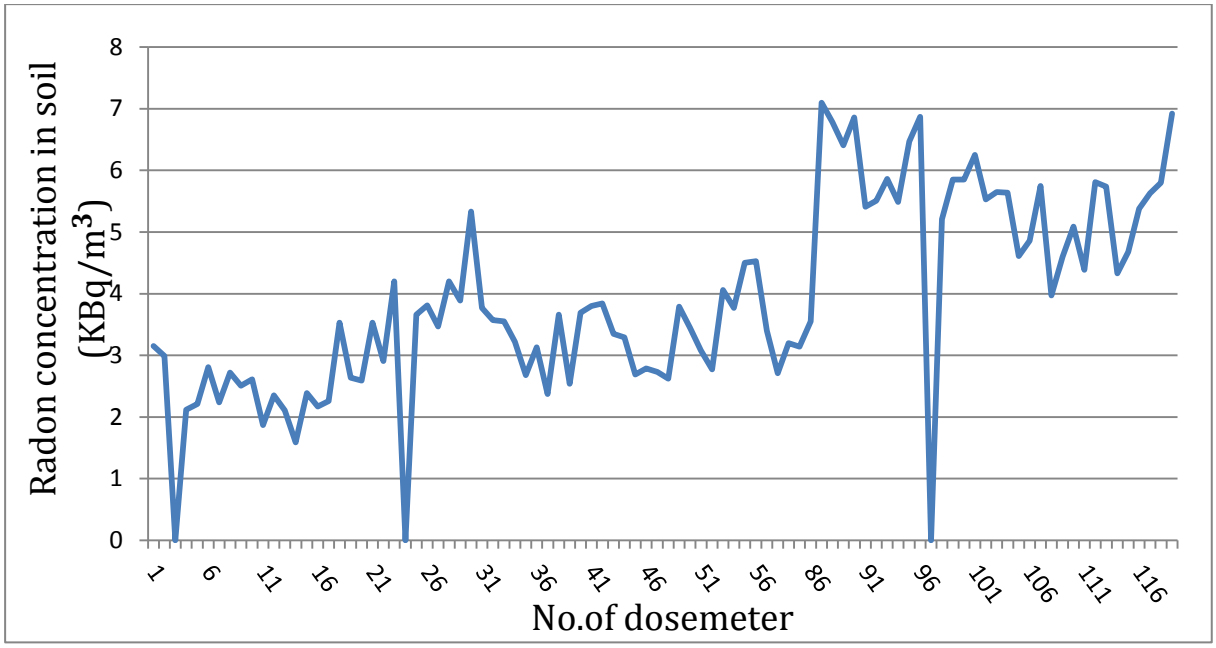

Fig.4-3: The radon concentration in soil at depth 50 cm, for horizontal study in the locations A and B.

4.2.2 Radon Concentration at Soil Surface (at depth of 0 cm) in Western Part of Location A

As displayed in Table 4-10, it is noticed that radon concentration is between (0.15 KBq m^{-3}) as the minimum concentration and (2.20 KBq m^{-3}) as the maximum concentration. The average concentration is 0.915 KBq m^{-3}.

The variations of radon concentration at the surface of soil are attributed to many factors, like the porosity and permeability of soil and the volume, form, and distribution of the soil's grains. Moreover, the moisture content and surrounding climate like temperature, atmosphere pressure and the amount of rain have a role in determining radon concentration at the surface of soil.

^{226}Ra is chemically similar to calcium and is absorbed from soil by plants. This leads to a decrease in the radon level at the surface and the layers just next to it [219].

4.2.3 Radon Concentration with Various Depths

As shown in the Fig.4-4, the concentration of radon in the soil air increases with depth, as noted in the Site A. The maximum radon concentration in depth 150 cm is 13.56 KBq m^{-3}. The minimum concentration at depth 10 cm is 0.78 KBq m^{-3}. In site B the maximum radon concentration at depth 80 cm is 11.91 KBq m^{-3}. The minimum concentration at depth 10 cm is 3.12 KBq m^{-3}.

When agricultural lands are irrigated, soil moisture increases in the upper layers, leading to a decrease in the radon concentration. Radon concentration increases as humidity decreases when going deeper into the earth, in addition to the effect of the atmospheric pressure which suppresses the diffusion of radon upward. There are some anomalous readings at both sites A, B and this might be caused by the different degrees of permeability and porosity.

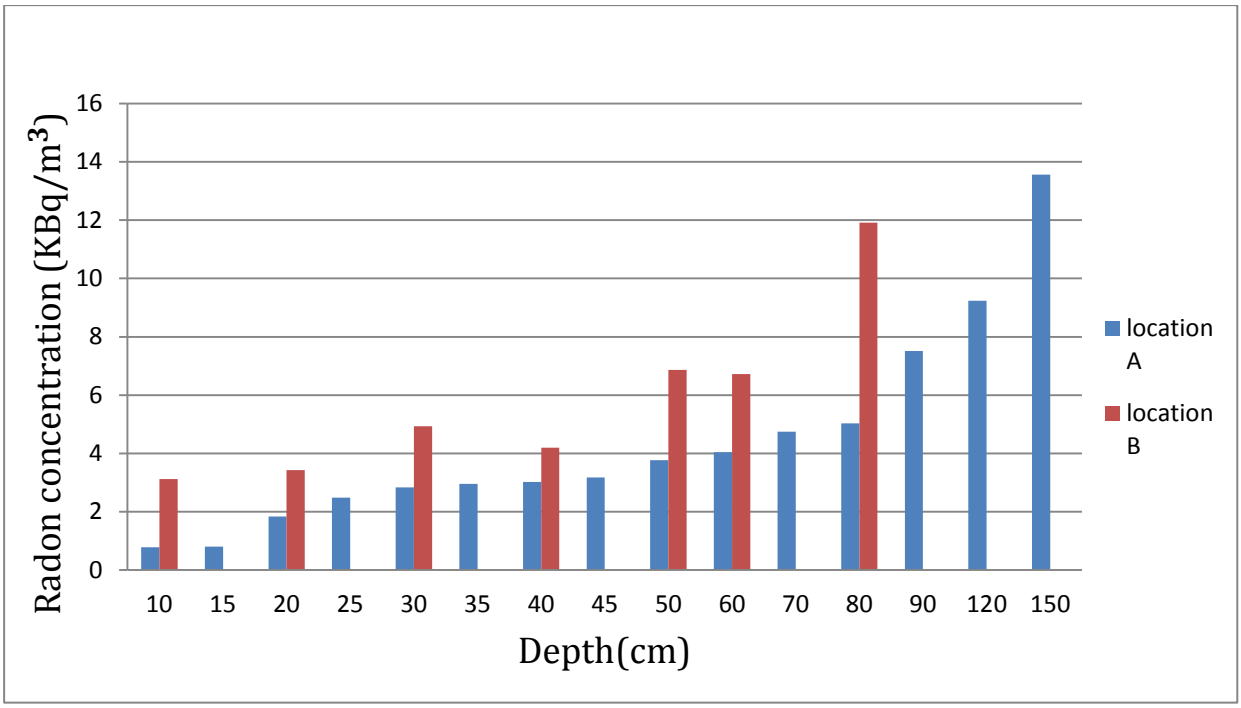

Fig.4- 4: Radon concentration in soil air at deferent depths.

Table 4-9: The average, minimum and maximum values of radon concentration in soil at depth 50 cm of horizontal for both locations A and B.

The radon concentration in soil at depth 50 cm (Horizontal study)	No. of dosimeters	Min. radon Conc. KBqm^{-3}	Max. radon Conc. KBq m^{-3}	Average radon Conc .KBq m^{-3}
The western part of the location A	(1-30)	1.59	5.33	2.728
The eastern part of the location A	(31- 60)	2.37	4.53	3.323
The south part of the location B	(86 - 102)	3.55	7.10	5.582
The north part of the location B	(103 - 119)	3.97	6.92	5.226
Total number of the four groups	94	1.59	7.10	3.887

Table 4-10: The average, minimum and maximum values of radon concentration with depth in soil and on soil surface at depth of (zero cm).

Type of the study	No. of dosimeters	Min. radon Conc. KBq m^{-3}	Max. radon Conc. KBq m^{-3}	Average radon Conc. KBq m^{-3}
The radon concentration with depth in location A	15	0.78	13.56	4.384
The radon concentration with depth in location B	8	3.12	11.91	5.146
The radon concentration on the soil surface (at depth of zero cm)	10	0.15	2.20	0.915

4.2.4 Conclusion and Recommendation

According to the results of radon concentration levels in the Table 4-9 and Table 4-10, the following points can be highlighted:

1- Results of the horizontal study of radon concentration in soil at depth of 50 cm: we found through the results that the average concentration of radon in the north part was approximately equal to the average concentration of radon in the south part in location B. Likewise, the average concentration of radon in the eastern part was approximately equal to the average concentration of radon in the western in location A. This convergence in both results is due to installation geological regions and method of tilling the soil and the type of fertilizer used.

2- The results of studying radon levels with depth indicate that the radon concentration increases up with depth to reach the maximum concentration. This may be due to the increase in pressure and moisture content with depth.

So when constructing in places of study, it should pay attention to the ground floors so that the repair of the existing cracks in the wall and the ground will be connected directly to the ground and thus will be more prone to leak and collect radon. The ground floors are to be equipped with a special ventilation system that constantly takes the radon out.

3- When the radon concentration in soil air is higher than 2000 Becquerel, and sufficient amounts of soil air leaked into homes, this may lead to the production of indoor radon gas higher than 300 Becquerel [266]. Therefore, the results obtained in all types of studies (except for the study of radon concentration at the surface) indicate that these levels of radiation are not safe and may pose a risk to human health, especially when houses built in the study area are poorly ventilated and the foundation on which these houses are built contains cracks from which radon can leak into these houses.

4.3. Measurement of Radon Exhalation Rates from Fertilizers

The average of radon concentrations in our samples is shown in Fig. 4-5. The highest radon concentration was found in sample F12, and the lowest radon concentration was found in Sample F9.

The values of radon concentration and exhalation rate of radon in the fertilizers samples, which were collected from different locations, are listed in Table 4-11.

As shown in Table 4-11, the radon concentrations range from 482.36 ± 36.519 to 1329.2 ± 30.446 Bq m^{-3}.

The study further found that the average concentration obtained in this study is 878.03 lower than the reported average of radon concentration in Saudi Arabia, which was 977.11 Bq m^{-3} [267].

The highest concentration of radon was in NPK fertilizers, which have phosphates in their production. Phosphate is known to contain large amounts of radioactive elements of natural origin. Additionally, the results show that radon concentration in organic fertilizers (animal manure) was higher than the radon concentration in nitrogen and potassium. This could be attributed to animals' food which contains natural radionuclides.

In addition, high levels of radon concentration in organic fertilizers may be explained by the animals' food which contains natural radionuclides.

As shown in Table 4-11, the surface and mass exhalation rates range from 400.62 to 1104 mBq m^{-2} h^{-1} and from 18.146 to 59.812 mBq kg^{-1} h^{-1}, respectively.

The effective radium content was found to vary from 1.967 to 5.722 Bq $kg^{-1}$with a mean value of 3.371Bq kg^{-1}.

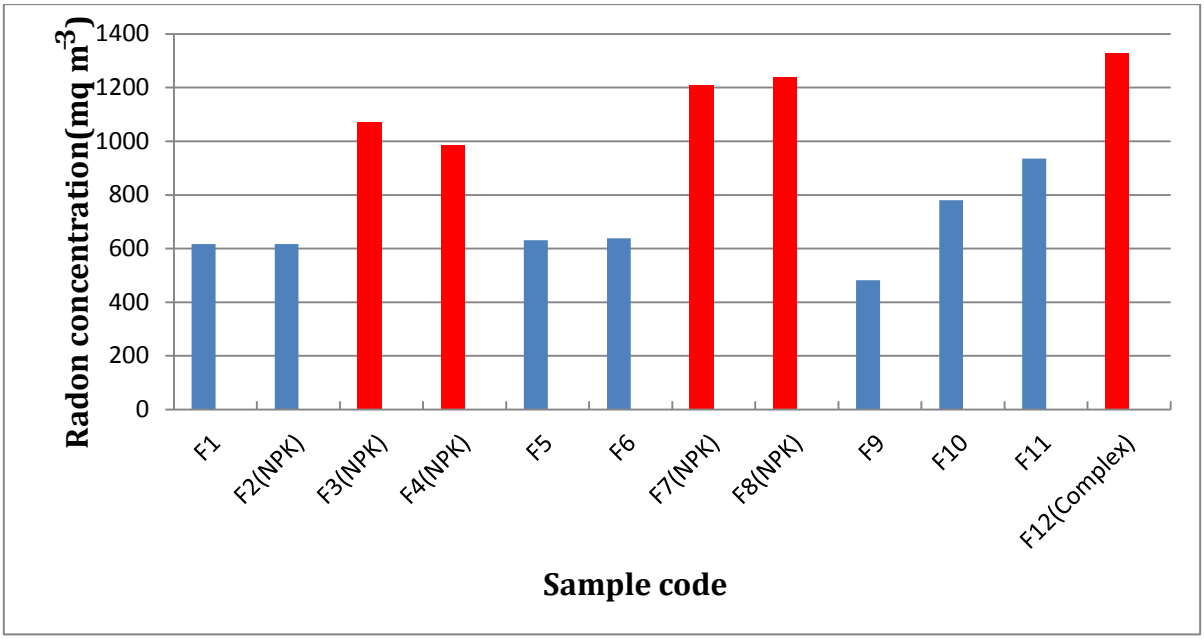

Fig.4-5: Concentration of radon in different fertilizer samples.

Table 4-12: Radon concentration, surface and mass exhalation rates and effective radium content for the different fertilizer samples.

Sample code	Type of fertilizer	Chemical composition	Radon Concentration (Bq m^{-3})	Radon surface exhalation rate (m Bq m^{-2} h^{-1})	Radon mass exhalation rate (mBq kg^{-1} h^{-1})	Effective radium content (Bq kg^{-1})
F1	Nitrogenous (Ammonium sulphate)	S=24, N=21	616.75±34.196	512.24 ±28.40	23.202±1.286	2.459
F2	Complex (NPK)	13/40/13	616.76±33.351	512.25±27.70	23.202±1.255	2.515
F3	Complex (NPK)	20/20/20	1069.7±45.055	888.40±37.42	40.239±1.695	4.362
F4	Complex (NPK)	5/5/5	984.96±44.689	818.06±37.12	37.053±1.681	3.794
F5	Potassic (Potassium sulfate)	S=18, K=51	631.48±34.133	524.48±28.35	23.756±1.284	2.804
F6	Nitrogenous Fertilizers(Urea)	N=46	638.84±36.458	530.59±30.28	24.033±1.372	2.316
F7	Complex Fertilizer (NPK)	3/37/37	1209.6±44.24	1004.6±36.74	45.503±1.664	5.152
F8	Complex Fertilizer (NPK)	N=20, P2O5=20 K2O=20	1240.9±42.245	1030.6±35.09	46.680±1.589	5.061
F9	Potassic (Potassium sulfate)	K2O=20, S=18	482.36±36.519	400.62±30.33	18.146±1.374	1.967
F10	Organic		780.61±27.189	648.33±22.58	49.922±1.739	1.996
F11	Organic		935.25±48.261	776.78±40.08	59.812±3.086	2.306
F12	Complex Organic-mineral	*COMPLEX	1329.2±30.446	1104.00±25.29	50.005±1.145	5.722
MAX			1329.2±30.446	1104.00±25.29	59.812±3.086	5.722
MIN			482.36±36.519	400.62±30.33	18.146±1.374	1.967
Average			878.03±38.065	729.25±0.032	36.795+1.597	3.371
*ORDNIC MATTER≥20%, SEAWEED EXTRACT ≥5, HUMICACID≥≥5, CaO≥10%, MgO≥10%, Fe≥5%						

4.3.1 Conclusion and recommendation

All of the values of radon concentrations in Nitrogenous and Potassic fertilizers were below the internationally accepted levels recommended by the International Commission on Radiological Protection (ICRP 2014) for work places, 1000 Bq m^{-3}.

Based on the results, it can be seen that organic fertilizer samples contain a high radon concentrations that are close to the internationally determined value by the (ICRP 2014)

The result from our survey shows that most NPK fertilizers sample and complex fertilizer sample contain a high radon concentrations that are higher than the internationally determined value of the ICRP2014 (see Fig. 4-5 and Table 4-1).

The use of these fertilizers should, when necessary, be made under strict supervision and inspection by the Ministry of Agriculture and the Ministry of Health, in order to avoid any health risks to farmers, farms, soil and animals.

Also, we suggest that fertilizers should be stored in warehouses away from offices and residences to minimize health hazards resulting from continuous exposure

Adequate ventilation is mandatory in fertilizer warehouses to limit exposure of workers to radon since radiation exposure is cumulative.

Using fertilizers every year is expected to cause detrimental effects with regard to radioactivity pollution and this should make it imperative to monitor changes that could occur in the soil and to develop pollution control strategies.

The results indicate that radon concentrations and exhalation rates significantly vary from one sample to another. This may be due to the difference in the chemical composition and the geological form of the samples, depending on whether the distribution of naturally occurring radionuclides present in soil and rocks is nonuniform in the earth's crust.

All the values of effective radium content in all samples under test were found to be quite lower than the permissible value recommended by the OECD [235].

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