UNIVERSITI PUTRA MALAYSIA

ASSESSMENT OF THE EFFECT OF RADIONUCLIDES FROM MALAYSIAN SOILS ON ENVIRONMENTAL SAFETY

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ASSESSMENT OF THE EFFECT OF RADIONUCLIDES FROM MALAYSIAN SOILS ON ENVIRONMENTAL SAFETY

by
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In the name of Allah, the Merciful, the Compassionate.

Praise be to God, Lord of the universe,
The Gracious, the Merciful,
Master of the Day of Judgment.
You alone we worship; You alone we implore for help.

Guide us unto the straight path-
The path of those whom You blessed,
Those who have not incurred Your displeasure,
Those who have not gone astray.

Amen
Qur'an, 1:1-7

First and foremost, I would like to express my sincerest thanks and appreciation to my supervisory committee chairman Prof. Dr. Haji Mohd. Yusof Sulaiman for his advice, guidance, useful suggestions, helpful discussions and most of all for his kindness throughout this study.

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No single word or expression of appreciation can adequately reflect my gratitude and love to my parents, my brothers, sisters and my wife Rahimah for their moral support and understanding, and to my child Siti Amirah for providing pleasant and comforting distraction.

An apple is red. The sun is yellow.
The sky is blue. A leaf is green.
A cloud is white ... and a stone is brown.

The world has many things ... the world has many people
the world has many colours ... and each of them is different.

In a garden
all the flowers
but they live happily together ... side by side.

In a forest
all the birds are different colours,
but they live happily together ... side by side.

In a meadow
all the animals are different
but they live happily together ... side by side.

In our world
all the people are different
and, sometimes, they live happily together ... side by side.

Colours are important
because they make our world beautiful,
but they are not as important as how we feel ...
or what we think ... or what we do.

Colours are outside things
and feelings are inside things.

Colours is something we see with our eyes,
but love is something we see with our heart.

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LIST OF ABBREVIATIONS

A	-	Atomic Weight
ADC	-	Analog to Digital Converter
ANSI	-	American National Standard of Institute
eV	-	Electron Volts
FEP	-	Full Energy Peak
FET	-	Field Effect Transistor
FWFM	-	Full Width at $1/50^{th}$ Maximum
FWHM	-	Full Width at Half Maximum
FWTM	-	Full Width at $1/10^{th}$ Maximum
GANAAS	-	Gamma Activity, and Neutron Activation Analysis System
HPGe	-	Hyper Pure Germanium
IAEA	-	International Atomic Energy Agency
IEC	-	International Electrotechnical Commission
IEEE	-	Institute of Electrical and Electronics Engineers
LET	-	Linear Energy Transfer
M	-	Mass of Electron
MBSS	-	Marinelli Beaker Standard Source
MCA	-	Multichannel Analyzer
N	-	Distribution Factor
Q	-	Quality Factor
Z	-	Atomic Number

Abstract of the thesis submitted to the Senate of Universiti Putra Malaysia in fulfillment of the requirements for the Degree of Master of Science.

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August 1997

Chairman : Prof. Mohd. Yusof Sulaiman. PhD

Faculty : Science and Environmental Studies

Natural radioactive sources are the major contributor to human radiation exposure. The accumulation of radioactivity in surface soil is of concern since growing vegetation will take up radionuclides through roots and incorporate them into variou parts of the plants. Study of the level of radioactivity is soil is also important because natural exposure represents a reference against which exposure to man-made sources may be compared, not only for standards-setting purposes. but also in epidemiological studies. However little information is available on the distribution of natural radionuclides in Malaysian surface soil.

Surface soil sample were collected from uncultivated, unfertilized sites in a few selected areas through out Peninsular Malaysia. The concentration of ^{226}Ra, ^{212}Pb and ^{212}Bi which are members of the ^{238}U Series and ^{228}Ac, ^{212}Pb and ^{208}Tl from ^{232}Th Series were measured. Radionuclides identification and its quantitative determination was obtained from direct measurement of gamma radiations using a gamma ray spectroscopy system. This consists of a 25% relative efficiency high purity germanium coaxial detector having a 1.67 keV resolution at 1.332 MeV and a peak to-compton ratio of about 60 for the ^{60}Co peaks. The data was collected using a PCA-II 8192 channel analyzer.

The gamma ray spectrum analysis, energy calibration, efficiency calibration and activity determination were performed using a computer routine called Gamma Activity and Neutron Activation Analysis System (GANAAS) procured from International Energy Agency (IAEA). For peak fitting, GANAAS uses peak shapes consisting of a Gaussian central peak and modified exponential tail functions. The peak shape functions are actually incoporated in a fitting routine code named GAMANAL in the GANAAS computer program. Due to the fact that the quantification of the activities of radionuclides using this technique is quite complicated because of the presence of interfering radiations with energy close to each other. the procedure used to account for this effect was throughly discussed.

The average concentrations of ^{214}Bi and ^{228}Ac in Bq/kg were 198 and 251, respectively. To calculate the dose delivered, models linking the measured amounts that are released by the radionuclides in soil samples with the resulting dose developed in the irradiated subjects (human being) were used.

Abstrak tesis ysng dikemukakan kepada Senat Universiti Putra Malaysia bagi memenuhi syarat untuk memperolehi Ijazah Master Sains.

PENILAIAN KESAN RADIASI SEMULAJADI DARIPADA TANAH DI MALAYSIA KE ATAS KESELAMATAN ALAM SEKITAR.

oleh
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Ogos 1997

Pengerusi : Prof. Mohd. Yusof Sulaiman, PhD

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Sumber radioaktif semulajadi merupakan penyumbang utama dedahan radiasi kepada manusia. Pertambahan bahan radioaktif di dalam tanah permukaan perlu diberi perhatian sewajarnya memandangkan ianya akan diserap oleh tanaman melalui akar dan kemudiannya disebarkan kepada bahagian yang lain. Kajian ke atas paras sinaran radioaktif semulajadi di dalam tanah permukaan juga penting kerana ianya dapat dijadikan perbandingan dengan dedahan sinaran buatan manusia. Ianya bukanlah sekadar bahan rujukan sahaja malah amat berguna terutamanya ketika berlakunya dedahan yang tidak terancang ataupun

ketika bencana nuklear. Namun begitu, amat kurang informasi ataupun data tentang taburan paras sinaran semulajadi di dalam tanah permukaan di Malaysia.

Sample tanah permukaan dari beberapa kawasan tanah terbiar sekitar Semenanjung Malaysia telah diambil untuk dijalankan kajian. Kadar kandungan ^{226}Ra, ^{212}Pb dan ^{212}Bi dari Siri pereputan ^{238}U dan kadar kandungan ^{228}Ac, ^{212}Pb dan ^{208}Tl dari Siri pereputan ^{232}Th telah diukur menggunakan teknik pengukuran secara lansung sistem spektroskopi gamma. Sistem ini menggunakan pengesan koaksial "Hyper Pure Germanium" berkejituan relative sebanyak 25% serta mempunyai resolusi 1.67 keV pada 1.332 MeV dan kadar puncak ke-compton sebanyak 60 bagi puncak-puncak kobalt-60. Segala data telah dikumpul menggunakan penganalisa PCA-II yang mempunyai sebanyak 8192 rangkaian.

Analisis spektrum gama, tentukuran tenaga, tentukuran kecekapan dan penentuan aktiviti telah dibuat menggunakan perisian komputer "GANAAS" iaitu singkatan kepada Gamma Activity and Neutron Activation Analysis System yang dibekalkan oleh Agensi Tenaga Bangsa-bangsa Bersatu (IAEA). Segala prosedur dan teknik pembetulan sistem ini telah dibincangkan secara mendalam dan terperinci memandangkan bahawa kaedah ini agak rumit berpunca daripada kehadiran sinar radiasi yang begitu rapat di antara satu dengan yang lain.

Kadar purata bagi ^{214}Bi dan ^{228}Ac di dalam unit Bq/kg adalah sebanyak 198 dan 251. Akhirnya, model yang menghubungkan penyebar dedahan dan penerimanya telah digunakan bagi mengira kadar dos yang diterima oleh manusia hasil daripada dedahan radioaktif semulajadi tanah permukaan.

CHAPTER 1

INTRODUCTION

Life on earth is continuously subjected to radiation of natural origin. Exposure is both external and internal, the former arising from cosmic radiation and radionuclides in the environment, the latter arising from radionuclides taken into the body by ingestion or inhalation (UNSCEAR, 1988).

Natural sources are the major contributors to human radiation exposure. Study of these sources is important for several reasons. Natural exposure represents a reference against which exposure to man-made sources may be compared, not only for standards-setting purposes, but also in epidemiological studies of the consequences of man-made sources or even of unusually concentrated natural sources in certain areas. The variability in natural exposure introduces uncertainty in the nature of the control population in epidemiological studies.

Naturally existing radionuclides not of cosmic-ray origin and not members of decay chains must have half lives comparable to the several billion-year age of the earth. These radionuclides are few and only two, ^{40}K and ^{87}Rb, result in significant portions of the dose rate in humans due to natural sources of radiation.

There are three decay chains, the Uranium (4n+2) series, Thorium (4n) series and Actinium (4n+3) series, which occur naturally and whose parent radionuclides necessarily have half lives comparable to the age of the earth. The fourth series, Neptunian (4n + 1) series, does not occur in nature because the half-life of its longest lived member is short compared with the age of the earth (Faw and Shultis, 1993). Two decay series, the Uranium and Thorium, contribute appreciably to human exposure to natural radiation. The two important series are shown schematically in Figs. 1 and 2. While all the members of a series are not likely to be in radioactive equilibrium in nature for chemical or physical reasons, members of a subseries are likely to do so.

To calculate the dose delivered by radiation sources to exposed populations it is necessary to use models linking the measured or calculated amounts of radioactive materials that are released by the source or that are present in the environment, with the resulting dose in the exposed subjects (Faw and Shultis, 1993).

In this thesis relevant information regarding theory of the ionizing radiation, environmental pathway model of radionuclide buildup in soil, experimental setup of gamma spectroscopy system, spectrum analysis and the sample preparation of environmental samples are treated and discussed. Finally, results of the analyses performed are presented and conclusions drawn using the data available.

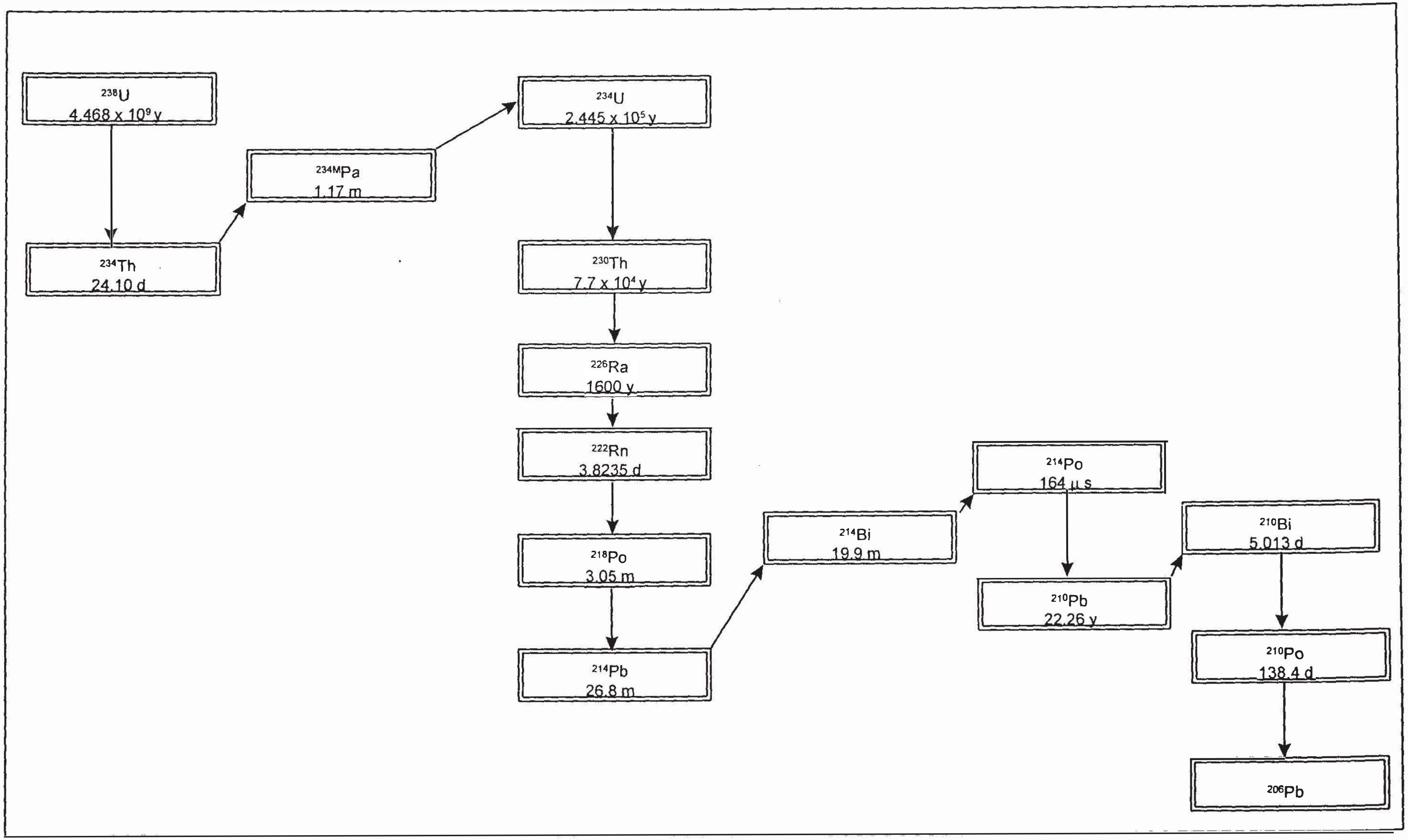

Fig. 1: Decay Scheme for ^{238}U.

Alpha Decay is depicted by vertically downwards lines and beta decay by lines upward to the right.

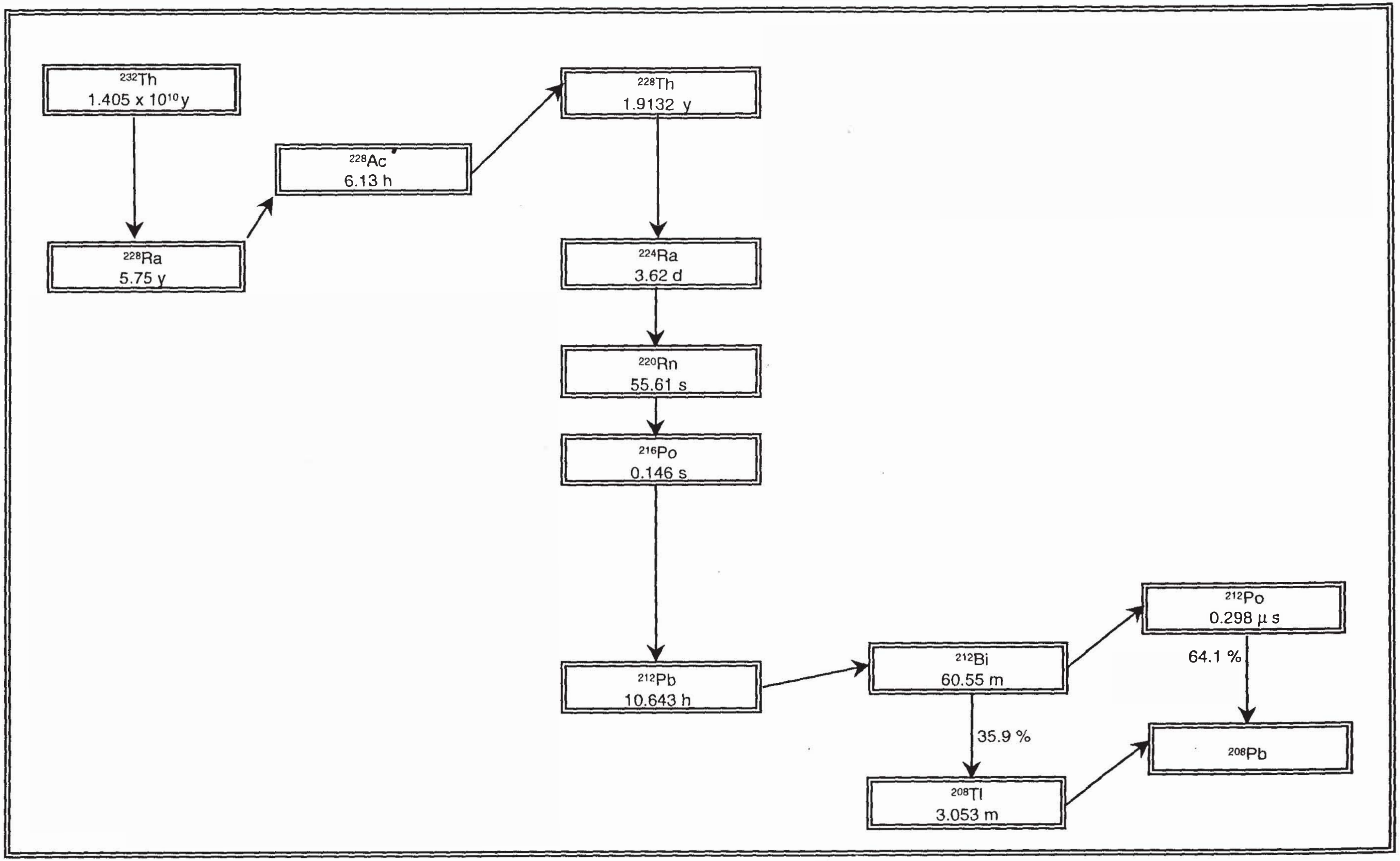

Fig. 2: Decay Scheme for ^{232}Th.

Alpha Decay is depicted by vertically downwards lines and beta decay by lines upward to the right.

CHAPTER II

PATHWAYS OF RADIONUCLIDES

Introduction

Radionuclides discharged into the environment can move through the biosphere by many routes that eventually lead to radiation exposure to humans. The movement of radionuclides along a particular pathway can be envisioned as proceeding through a series of pathway steps. Each step or compartment of a pathway represents some physical entity or process by which radionuclides are received from a donor compartment, accumulated, and eventually passed down the pathway to the next compartment. At the beginning of the pathway is the source of radioactivity while at the end of the pathway is the human population of concern whose internal exposure from ingestion of radionuclides or external exposure from radionuclides in the environment is to be estimated.

The exposure pathways that generally are the most important for humans are those in the terrestrial ecosystem. Two types of terrestrial exposure pathway are responsible for most of the population exposure. First, the accumulation of radionuclides on the ground leads to direct external exposures. Second, the contaminated leads to internal exposures.

Radionuclide Buildup in Soil

The accumulation of radioactivity in soil near the surface is of concern since growing vegetation will take up radionuclides through roots and incorporate them into various parts of the plants. Soil contamination thus represents a long-term source of radionuclides that can continue to contaminate human or animal food crops for many years after the initial soil deposition. By contrast, the direct contamination of vegetation is usually of concern only for the crops exposed during the deposition.

Soil can become contaminated from the direct deposition of airborne radioactivity, from contaminated rain or irrigation water, from the washoff of radionuclides initially deposited on plants, and from the decay of contaminated vegetation. All soil contamination initially enters the soil from the surface. These radionuclides then migrate into the soil, carried down by rain and irrigation water or mixed mechanically by tillage. The radioactivity available for subsequent uptake by plants is limited to that residing within the plant root zone, for most crops to a depth of 0.15 to 0.2 m. Radionuclides deposited on the ground can be removed from potential plant uptake by being washed away from the surface by surface water runoff or by recolation to a depth beyond the root zone brought about by water infiltration or by deep tillage.

Calculation of Soil Concentrations

For calculation of plant uptake at some location, one must first determine the average concentration $X_{soil}(t)$ of radioactivity (per unit mass of soil) in the root zone. This concentration is given by a simple balance relation between the soil input rate I and loss rates by decay (λX_{soil}) and by migration out of the root zone $(\lambda_s X_{soil})$, namely

$$\frac{dX_{soil}(t)}{dt} = I(t) - (\lambda + \lambda_s) X_{soil}(t). \qquad [2.1.]$$

Thus, for a constant input I_0 into the root zone beginning at time $t = 0$ and for a constant infiltration loss rate λ_s, the activity concentration in the soil is

$$X_{soil}(t) = \frac{I_0}{\lambda + \lambda_s}(1 - e^{-(\lambda + \lambda_s)t}). \qquad [2.2]$$

Let the rate at which a given type of radionuclide is deposited on a unit area of the surface at time t be denoted by $\omega_s(t)$. A fraction f_r of this surface deposition will be carried away by rain or irigation water that runs off the field, and the remainder $(1-f_r)$ will begin to migrate into the soil. Thus, the rate at which radionuclides enter the soil per unit mass of the soil, and averaged over the soil to depth d of of the root zone, is

$$I(t) = \frac{(1 - f_r)\omega_s(t)}{\rho d}, \qquad [2.3]$$

where ρ is the soil mass density (typically 1.6 to 2.6 g cm^{-3}). If only direct deposition to the ground from airborne radionuclides is considered, then the rate of direct input to the soil becomes

$$I(t) = (1 - f_{r)}[(1 - f_{veg})v_d + (1 - f_w)W_v R_p]\frac{X_{air}(t)}{\rho d} \qquad [2.4]$$

Here f_{veg} and f_w are the fraction of radionuclides removes from the atmospheric plume by dry and wet deposition on the vegetation, respectively, v_d is the dry deposition speed, W_v is the volumetric factor, R_p is the preprecipitation rate and $X_{air}(t)$ is the activity concentration in the air. This result neglects the subsequent input of radionuclides initially deposited on the vegetation but subsequently weathered or washed off the plants onto the ground. To account for plant washoff, Peterson (1983) suggests multiplying this result by the factor $[1 + k_{rain}R_p / (\lambda + k_{rain}R_p)$ where λ is the radioactive decay constant, and k_{rain} is an empirical constant. For long-term studies of soils in which the leafy material is allowed to decay back into the soil, the soil gain rate may be approximated by Eq.2.4 with the plant retention factors f_{veg} and f_w set to zero.

The fraction f_r of the surface deposition that is washed away by the runoff of surface water depends on how quickly the soil can absorb rain or irrigation water as well as on slope of the field. The capacity of soil to absorb water is measured by the soil permeability μ_{soil} which is defined as the water volume transmitted per unit surface area per unit time. The permeability varies between 2.5 to 25 cm h^{-1} for sandy soils, 1 to 7.5 cm h^{-1} for loam, and 0.025 to 0.5 cm h^{-1} for silt and clay. Thus, if the rainfall (or