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Evaluation of the contributions of ^{232}Th and its progenies to the ingestion dose coefficients

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Abstract

In International Commission on Radiological Protection (ICRP) Publication* 69*, a dosimetric multcompactmental model for ^{232}Th was proposed for possible application and testing its reliability in the estimation of dose to critical cells of the body. The present work employed this dosimetric model as proposed by the ICRP to estimate the ingestion dose coefficients to organs/tisues of the body using thorium/daughter hybrid method with serial transformation to accumulate nuclear transformation. This method made it possible to estimate the contributions of ^{232}Th and its progenies to the ingestion dose coefficients. The results obtained revealed that the estimated ingestion dose coefficients compared very well with the ICRP values. Moreover, the alpha emitters in the thorium chain contributed more than* 95% *to the dose coefficients. It is also believed that the present effort has contributed to the validation of the ICRP model for possible application to similar radioelement.

Keywords
Model, thorium, progenies, intake, dose coefficients

1.0 Introduction

Thorium, along with it progenies, emits six alpha particles and is therefore considered as one of the highly radiotoxic elements. ^{232}Th (half-lives of 1.4 x 10^{10}y) as the parent radionuclide decays with the emission of an alpha particle to ^{228}Ra (5.75y). The other decay modes leading to the emission of the other alpha particles involve the decay of ^{228}Th (1.91y) to ^{224}Ra (3.66d); ^{224}Ra (3.66d) to ^{220}Rn (55.5s); ^{220}Rn (55.5s) to ^{216}Po (0.15s); ^{216}Po (0.15s) to ^{212}Pb (10.6h); ^{212}Bi (60.6m) to ^{208}Tl (3.10m). The average activity concentration of ^{232}Th is in the range 25 – 50 Bq kg^{-1} and can concentrate in certain rocks like granites and alkaline igneous rocks [1]. The health hazards associated with these radionuclides stem from their ability to accumulate in human tissues, especially bone surface [2]. During the decay process, highly penetrating gamma rays are emitted, thereby causing intensive damage to the tissues where they are localized.

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The dose delivered to any organ of the body after ingestion or injection of a radioactive parent is a function of the radioactive half-lives, chemical compound form, and metabolism. In most cases, the metabolism of the parent nuclide may be somewhat different from that of its radioactive progenies within the decay series. Moreover, a progeny born in an organ through the decay of its radioactive parent may metabolically behave differently from that deposited directly through translocation from blood. For example, ^{228}Ra deposited directly from blood will behave as a bone volume seeker rather than that born in bone surface through the decay of its radioactive parent ^{232}Th. In view of the strong correlation between the metabolism of the radioactive progeny and the estimation of the doses delivered to a tissue or organ of the body, understanding the behaviour of radioactive progenies after intake of a radioactive parent is important for proper estimation of such doses. Unfortunately, there are no experimental human data on the metabolism of progeny born in various organs and tissues [3]. According to this authors, it is not likely whether the metabolism of the parent can be apply to its progeny and that it is more probable for the progeny to follow the metabolic behaviour of their own element. ^{232}Th is a bone surface seeker when introduced in the body [4]. The International Commission on Radiation Protection [5] described the metabolism of metabolites and their retention in specific organs in the body using multicompartmental model. In the model, it was assumed that radioactive progeny born from the decay of ^{232}Th in the body metabolize as their ^{232}Th precursor. According to Dewhey et al [3], this assumption was made to simplify the mathematics in calculating the number of transformations in and the doses to target tissues. The radioactive element ^{228}Ra is known to behave like its chemical analogue calcium, which is a major element within mineral bone. It is most probable for it to be incorporated within bone volume and will likely behave metabolically as alkaline earth elements during bone remodelling process.

In this work, the contributions of the various progenies of thorium to the dose coefficients in the different organs and tissues of the body normally employed to calculate doses to these organs and tissues were estimated using the biokinetic and dosimetric models for ^{232}Th described by ICRP [6] that assume same metabolic behaviour for both radioactive parent and progeny. In publication 69 of ICRP [6], the effective dose coefficients in different organs and tissues for six age groups have been compiled for radiation protection of members of the public and workers. However, this age – dependent dose coefficients were not given explicitly to include the contributions of the individual radioactive daughters in the thorium decay chain. The present work will provide information on which of the progenies could be included in the establishment of more reliable estimates of doses from thorium. The reliability of the accumulated nuclear transformation method employed in this work will elicit a novel approach to the evaluation of dose coefficients for other radionuclides.

2.0 Theory

2.1 Biokinetic modelling of thorium and its progenies

Radionuclides transport in the human body can be investigated using deterministic model. This process involves model simulation of the linear transfer processes represented by sets of linear differential equations governed by first order kinetics. Given the compartmental model of the GI section of Fig 1 for example, if $q(t)$ is the radionuclide activity ingested in a compartment at time t, the model is described by the following equations:

$$\frac{d}{dt}q_{ST}(t) = -\lambda_{ST}q_{ST}(t) - \lambda_R q_{ST}(t) + \dot{I}(t) \qquad (2.1)$$

$$\frac{d}{dt}q_{SI}(t) = -\lambda_{SI}q_{SI}(t) - \lambda_R q_{SI}(t) + \lambda_{ST}q_{ST}(t) - \lambda_B q_{SI}(t) \quad (2.2)$$

$$\frac{d}{dt}q_{ULI}(t) = -\lambda_{ULI}q_{ULI}(t) - \lambda_R q_{ULI}(t) + \lambda_{SI}q_{SI}(t) \quad (2.3)$$

$$\frac{d}{dt}q_{LLI}(t) = -\lambda_{LLI}q_{LLI}(t) - \lambda_R q_{LLI}(t) + \lambda_{ULI}q_{ULI}(t) \quad (2.4)$$

where λ_R is the radioactive decay constant of the radionuclide in question

$\lambda_B q_{ST}(t)$ is the rate of transfer of activity to the body fluid (systemic circulation)

λ_i is the biological clearance rate from the compartment *i* to the other

$\lambda_i q_i(t)$ is the rate of transfer of activity from the compartment *i* to the other

$\lambda_R q_i(t)$ is the decay rate of the radionuclide activity in the compartment *i*

$I_i(t)$ is the rate of intake of the activity from outside into the compartment *i* at time *t*.

To model these multi-compartmental systems and to solve the sets of linear differential equations, different software packages are available for solving multi-compartmental systems. Skrable et al [7] developed a blood-organ transfer kinetics model for solving first order compartmental models. Johnson and Myers [8] applied the general model by Skrable et al [7] to obtain retention equation for the alkaline earth elements. This model was later improved upon by Spacher [9] by the addition of one compartment and a more detailed description of the systemic excretion pathways was given. Dewhey et al [3] then proposed metabolic models for serial decay of ^{232}Th/^{228}Ra/^{228}Th/^{224}Ra by using Skrable et al [7] blood-organ transfer kinetics to develop a thorium metabolic model and then coupled it with Spacher's radium model to estimate the total committed dose equivalent per unit intake of ^{232}Th, ^{228}Ra, ^{228}Th, and ^{224}Ra.

In the present work, the metabolic model for ^{232}Th (Figure 2.1) and its progenies given by the ICRP was employed in the estimation of the contributions of the various members of the ^{232}Th decay chain to the ingestion dose coefficients and the effective dose. To achieve this, hybrid method involving nuclear transformation of content in each compartment into the corresponding compartment of the next radioactive daughter was employed (Figure 2.2). The content received from the precursor radioactive nuclide serves as initial input for the immediate progeny. The process goes on in the integral time course up to the end of the decay chain using their individual radioactive half-lives as the transfer kinetics.

In the compartmental model shown in Figure 2.1. The pathways of radionuclide transfer between the compartments are depicted by the arrows shown in the figure. The transfer rates between compartments given by the ICRP [6] for adult were used for the radionuclide in the chain. These transfer kinetics were fed into the model setup of the SAAM II module as shown in Figure 2.2. The model is solved by accumulating nuclear transformation in each compartment for each of the radionuclide in the chain. In the estimation of the accumulated transformations in any compartment of interest in the hybrid metabolic model with serial transformation, additional compartments were added to the model compartment to accumulate disintegration for the radionuclide under consideration.

For the purpose of simulating the behaviour of the radionuclides between compartments after ingestion, the systemic model was coupled to the gastrointestinal (GI) tract model. The transfer rate from the small intestine to blood was calculated according to the appropriate absorption fraction (f_1 value) for each progeny given by ICRP Publication 69 [6].

2.2 Estimation of internal dose

In this work, the age-dependent and dosimetric formulations as described by Eckerman [10] were adopted. The average organ dose, D_T, in an organ or tissue *T*, due to nuclear transformations of a radionuclide in various source organs *S*, is given as:

$$D_T = \sum_s D(T \leftarrow S) \tag{2.5}$$

where $D(T \leftarrow S)$ is the absorbed dose in a target organ or tissue (T) from a radionuclide in a

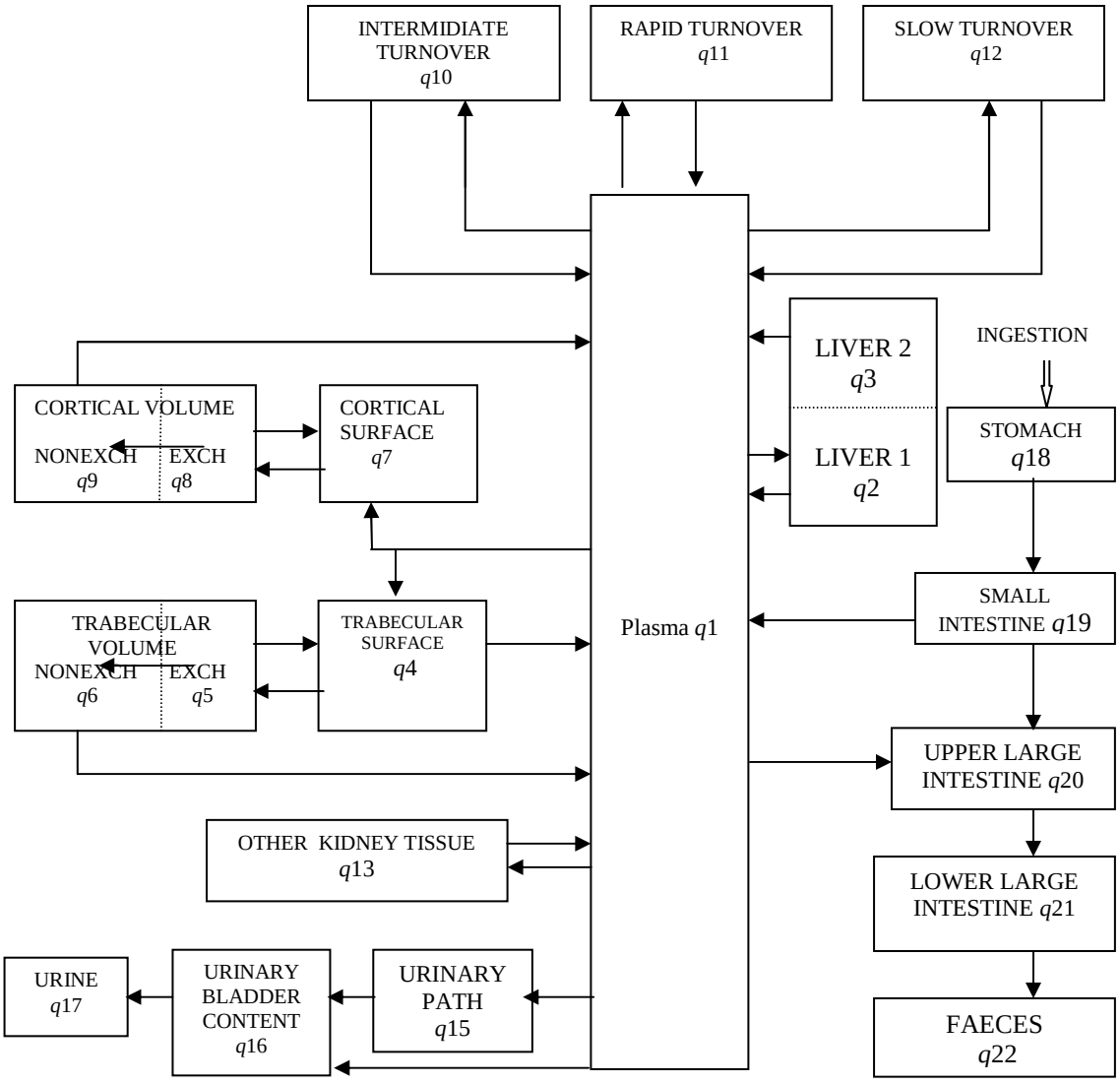

Figure 2.1: Compartmental model for thorium as recommended by the ICRP [6]

single source organ or tissue (S). The absorbed dose is given as:

$$D(T \leftarrow S) = U_s \times c \sum_i \frac{E_i \times Y_i \times \Phi_i (T \leftarrow S)}{M_T} \tag{2.6}$$

where U_S is the cumulated number of nuclear transformations of the nuclide in S; c is a numerical constant required by the units of the associated quantities (for E_i in J, M_T in kg and $D(T \leftarrow S)$ in Gy per nuclear transformation, $c = 1$); E_i and Y_i are the mean energy and the yield

of radiation type i, respectively; $\Phi_i(T \leftarrow S)$ is the absorbed fraction of energy of radiation type *i*; and M_T is the mass of the target organ, *T*.

The equivalent dose, H_T, is given by summing all source organs, *S*, and radionuclide, *j*, as:

$$H_T = c\sum_s \sum_j U_{sj} \times SEE\left(T \leftarrow S\right)_j \tag{2.7}$$

where

$$SEE\left(T \leftarrow S\right)_j = \sum_i \frac{E_i \times Y_i \times W_R \times \Phi_i\left(T \leftarrow S\right)}{M_T} \tag{2.8}$$

is the specific effective energy deposited in target *T* per nuclear transformation of radionuclide *j* in source *S*. w_R is the radiation weighting factor.

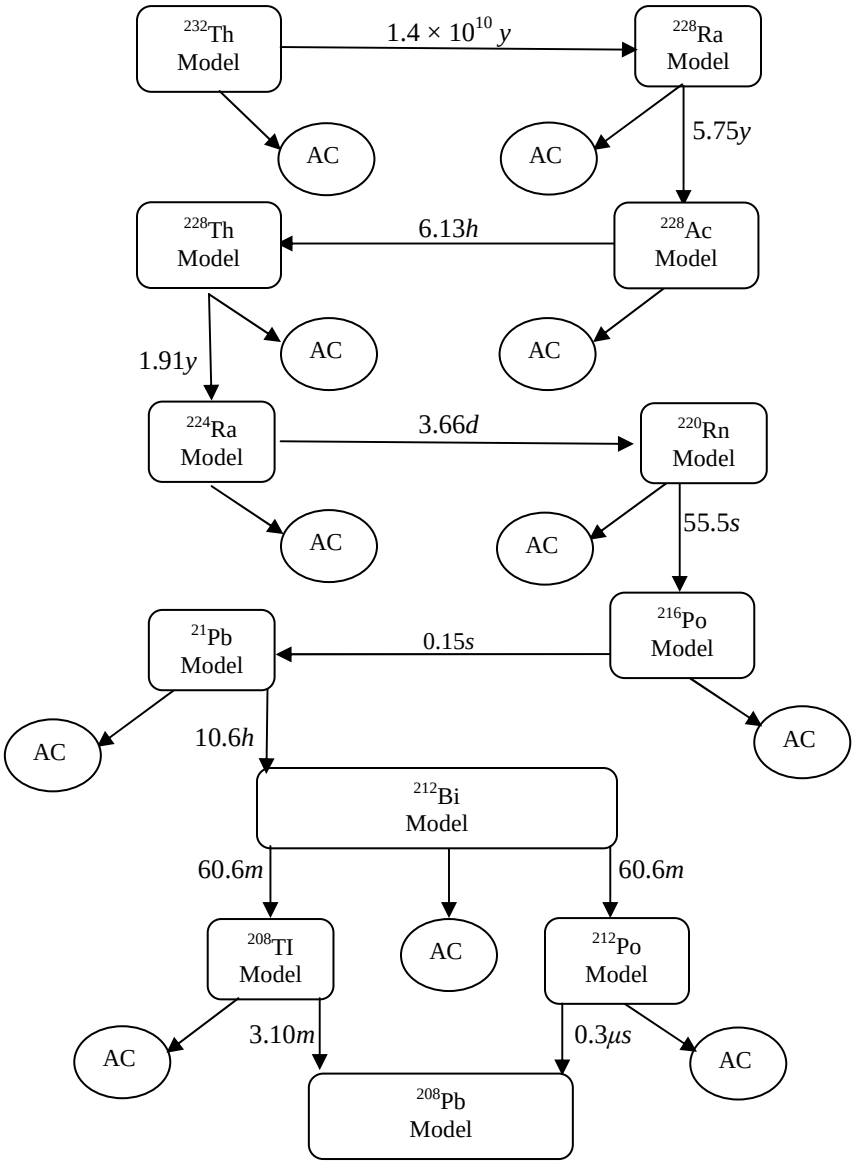

Figure 2.2: Hybrid compartmental model for thorium and progenies using the physical half-live of each radionuclide to transfer contents from the compartments represented in fig 1 to the adjoining radionuclide. An additional compartment (**circle AC**) was added to each model compartment to accumulate disintegration using the individual radionuclide half-live as the transfer rate.

The accumulated nuclear transformations, U_S, for the various radionuclides in the thorium decay chain were obtained by bolus ingestion of 1*Bq* using the model set up given in Figure 2.2. The age-dependent $SEE(T \leftarrow S)_j$ were calculated using the software SEECAL [11, 12] developed by Oak Ridge National Laboratory (ORNL). The effective dose, E, was calculated according to ICRP [6, 13] as:

$$E = \sum_T W_R \times H_T + R_{remainder} H_{remainder} \tag{2.9}$$

where $H_{\text{remainder}}$ and $w_{\text{remainder}}$ are the equivalent dose and the weighting factor for the remainder tissues. The weighting factors for target cells in different lung region recommended by the ICRP [14] were used in the estimation of the contribution from the lung.

3.0 Results and discussion

The accumulated transformations from the various model simulations for thorium and its progenies were tabulated in a database in MS Excel format. The dose equivalent (H_T) is calculated using equation (2.3), where the specific effective energy (SEE) deposited in target organ *T* per nuclear transformation of radionuclide in source organ *S* was calculated using equation (2.4). The result obtained for the committed dose equivalent and the effective dose calculated using equation (2.5) as compared with the ICRP values is listed in Table 3.1. The % difference and the ratio between the ICRP values and this work are also listed in the table. The result shows that the % difference is less than 3% for all the organs/tissues except for the testes that is about 9%. This higher percentage recorded for the testes as against the other organs/tissues is due the fact that the present work applied the transfer rate from blood to the testes by excluding that for the female organ. The model is such that the transfer of contents in and out of the gonad compartment can only be described by either transfer rates.

The percentage contributions of the various radionuclide daughters of thorium to the committed dose equivalent in the various organs or tissues are presented in Table 3.2. The table reveals that the alpha emitters in the decay chain contributed more than 95% to the committed dose equivalent. ^{232}Th contributed 88% to the bone surface, this is expected since it is a bone surface seeking radionuclide. Significant contributions in other organs like the lower large intestine wall, kidney, liver, red marrow and testes can be observed. Hence, it contributed about 80% to the effective dose. The implication of this is that the effective dose can only be underestimated by about 20% when ^{232}Th is considered without its progenies. However, it has to be noted that 20% is very significant in the estimation of dose to the internal organs. Hence, the contributions of other alpha emitters in the decay chain must be taken into account in order to have reliable internal dose estimates from the dietary thorium uptake into the body circulation. The high contributions from alpha emitters in the thorium decay chain to the dose delivered to the organs is due to the alpha energy recoil effect and their ability to ionize localised cell. For instance, ^{228}Ra nucleus is formed with a recoil energy of about 60 – 70 keV. This recoil energy is sufficient to break any chemical bonds in tissues and organs in the body [3]. Although the contributions from non-alpha emitters of the decay chain to the effective dose is less than 2%, ^{212}Bi percentage contribution in the group is about 50%. Among the beta emitters in the chain, it has a short half-live of an hour, which makes it disintegrate very fast to make significant contribution in most organs.

4.0 Conclusion

In this study, dose coefficient to the critical organs of the body has been estimated using the hybrid ^{232}Th model with its radioactive daughters. The contributions of its radioactive progenies to the committed dose equivalent have been estimated. The dose coefficients calculated in this present method compared well with the ICRP [6] values. The percentage difference is well below 3% for all critical organs/tissues except for the testes which is about 9%. The contribution from alpha emitters in the thorium decay chain to the effective dose to critical cells of the body is about 98%. Significant contribution (about 20%) from other alpha emitters apart from ^{232}Th has been noted. In addition, the present method has contributed to the
establishing the efficacy of the ICRP dosimetric model for thorium and also to the testing for possible application to other similar radioelement.

Table 3.1: Comparison between this work ingestion dose coefficient values and the ICRP values in the various organs of the body.

Organ/Tissue	Dose Coefficient (Sv/Bq)		%Difference	Ratio
	ICRP[6]	This Work		
Adrenals	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Urinary Bladder Wall	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Bone Surface	1.20×10^{-5}	1.20×10^{-5}	0.00	1.00
Brain	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Breasts	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Stomach Wall	3.80×10^{-8}	3.80×10^{-8}	0.00	1.00
Small Intestine Wall	4.01×10^{-8}	4.01×10^{-8}	0.00	1.00
Upper Large Intestine _Wall	5.20×10^{-8}	5.20×10^{-8}	0.00	1.00
Lower Large Intestine Wall	8.10×10^{-8}	8.10×10^{-8}	0.00	1.00
Kidneys	1.80×10^{-7}	1.80×10^{-7}	0.00	1.00
Liver	1.80×10^{-7}	1.80×10^{-7}	0.00	1.00
Lung	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Muscle	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Pancreas	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Red Marrow	4.60×10^{-7}	4.61×10^{-7}	0.33	1.00
Skin	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Spleen	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Testes	1.00×10^{-7}	0.91×10^{-7}	8.63	0.91
Thymus	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Thyroid	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Uterus	3.60×10^{-8}	3.70×10^{-8}	2.78	1.03
Remainder	3.70×10^{-8}	3.80×10^{-8}	2.70	1.03
Effective Dose	2.30×10^{-7}	2.29×10^{-7}	0.45	0.99

Table 3.2: Percentage contributions of radioactive daughters in the thorium decay chain.

Organ/ Tissue	% Contributions in the decay Chain					
	Th232*	Ra228	Ac228	Th228*	Ra224*	Rn220*
Adrenals	30.8	1.8×10^{-3}	0.31	10.6	11.0	12.3
Urinary Bladder Wall	30.9	1.8×10^{-3}	0.16	10.7	11.0	12.3
Bone Surface	88.7	0.3×10^{-3}	0.01	2.8	1.70	1.8
Brain	30.8	1.8×10^{-3}	0.26	10.6	11.0	12.3

Organ/ Tissue	% Contributions in the decay Chain					
	Th232*	Ra228	Ac228	Th228*	Ra224*	Rn220*
Breasts	30.9	1.8×10^{-3}	0.15	10.7	11.0	12.3
Stomach Wall	32.6	1.8×10^{-3}	0.16	10.4	10.8	12.0
Small Intestine Wall	34.9	1.7×10^{-3}	0.20	10.1	10.8	11.6
Upper Large Intestine Wall	49.0	2.4×10^{-3}	0.16	7.6	8.2	9.2
Lower Large Intestine Wall	65.3	3.3×10^{-3}	0.18	4.9	5.6	6.2
Kidneys	86.4	8.7×10^{-6}	0.03	2.6	0.6	0.8
Liver	73.6	4.9×10^{-4}	0.04	3.8	4.1	4.3
Lung	30.9	1.8×10^{-3}	0.20	10.8	11.0	12.3
Muscle	30.9	1.8×10^{-3}	0.21	10.8	11.0	12.3
Ovaries	30.9	1.8×10^{-3}	0.24	10.8	11.0	12.3
Pancreas	30.9	1.8×10^{-3}	0.22	10.8	11.0	12.3
Red Marrow	78.8	2.4×10^{-3}	0.10	5.9	3.0	3.3
Skin	30.9	1.8×10^{-3}	0.17	10.8	11.0	12.3
Spleen	30.9	1.8×10^{-3}	0.18	10.8	11.0	12.3
Testes	94.9	9.6×10^{-6}	0.04	1.7	0.4	0.8
Thymus	30.9	1.8×10^{-3}	0.18	10.8	11.0	12.3
Thyroid	30.9	1.8×10^{-3}	0.21	10.8	11.0	12.3
Uterus	30.9	1.8×10^{-3}	0.19	10.8	11.0	12.3
Remainder	33.6	1.7×10^{-3}	0.20	10.8	10.5	11.7
Effective Dose	80.4	9.9×10^{-4}	0.06	4.3	2.9	3.2

Table 3.2: Percentage contributions of radioactive daughters in the thorium decay chain. (contd)

Organ/ Tissue	% Contributions in the decay Chain					
	Po216*	Pb212	Bi212	Po212*	Tl208	Alpha emitters
Adrenals	13.3	0.050	4.26	16.89	0.370	94.89
Urinary Bladder Wall	13.3	0.040	4.28	17.00	0.200	95.20
Bone Surface	1.9	0.003	0.58	2.33	0.006	99.26
Brain	13.3	0.050	4.26	16.92	0.310	94.92
Breasts	13.3	0.030	4.26	16.97	0.160	95.17
Stomach Wall	12.9	0.030	4.16	16.55	0.180	95.25
Small Intestine Wall	12.5	0.040	4.01	15.95	0.240	95.85
Upper Large Intestine Wall	9.8	0.050	3.16	12.37	0.210	96.17
Lower Large Intestine Wall	6.7	0.080	2.28	8.44	0.260	97.14
Kidneys	0.8	0.010	1.73	6.88	0.060	98.08
Liver	4.7	0.010	1.88	7.52	0.050	98.02
Lung	13.3	0.040	4.26	16.94	0.240	95.24
Muscle	13.3	0.040	4.26	16.94	0.250	95.24
Ovaries	13.3	0.050	4.26	16.93	0.280	95.23
Pancreas	13.3	0.040	4.26	16.93	0.260	95.23
Red Marrow	3.5	0.020	1.06	4.05	0.080	98.55
Skin	13.3	0.030	4.26	16.96	0.190	95.26
Spleen	13.3	0.040	4.26	16.95	0.210	95.25
Testes	0.9	0.006	0.22	0.86	0.058	99.56
Thymus	13.3	0.035	4.26	16.96	0.200	95.26

*Alpha emitters

Table 3.2: Percentage contributions of radioactive daughters in the thorium decay chain. (contd)

Organ/ Tissue	% Contributions in the decay Chain					
Thymus	13.3	0.035	4.26	16.96	0.200	95.26
Thyroid	13.3	0.040	4.26	16.94	0.240	95.24
Uterus	13.3	0.040	4.26	16.95	0.230	95.25
Remainder	12.7	0.038	4.14	16.44	0.240	95.74
Effective Dose	3.5	0.014	1.10	4.31	0.050	98.61

*Alpha emitters

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