A NEW SIMULATION MODEL FOR CALCULATING THE INTERNAL EXPOSURE OF SOME RADIONUCLIDES

by

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A new model based on a series of mathematical functions for estimating excretion rates following the intake of nine different radionuclides is presented in this work. The radionuclides under investigation are: cobalt, iodine, cesium, strontium, ruthenium, radium, thorium, plutonium, and uranium. The committed effective dose has been calculated by our model so as to obtain the urinary and faecal excretion rates for each radionuclide. The said model is further validated by a comparison with the widely spread Mondal software and a simulation program. The results obtained show a harmony between the Mondal package and the model we have constructed.

Key words: radionuclides, Mondal package, simulation

INTRODUCTION

Occupational exposure to ionizing radiation can occur in many industries, such as mining, medical institutions, educational and research establishments, and nuclear fuel facilities. Adequate radiation protection of the workers is essential for a safe and acceptable use of radiation, radioactive materials and nuclear energy [1].

Internal exposures occur when radionuclides are inhaled, ingested, or otherwise absorbed by the body through wounds and intact skin. A proportion of the inhaled material will eventually be swallowed; in that case, the radionuclides inside the body are called internal emitters [2].

Individual monitoring for internal exposure is based on the direct measurement of radionuclides in ex-

creta. The removal of deposited material from the body, in principle, occurs through urinary and fecal excretion [3, 4]. The biological samples used for the estimation of intake and assessments of internal exposure are, most commonly, urine and feces, although breath and blood can also be used in special cases [5, 6].

In the last ten years, the ICRP (International Commission on Radiological Protection) has revised the human respiratory tract models [7]. These developed models permit a more realistic description of the behavior of radionuclides in the human body, including its excretion processes. Griffith *et al*. [8] reviewed a simple bioassay model for predicting the organ burden of [241]Am from the excretion rate presented for inhalation exposures. The model uses three compartments representing the lungs, liver, and skeleton. It was developed using data from studies of inhaled or injected [241]Am in laboratory animals and validated for comparison with people in cases of accidental inhalation exposures to [241]Am.

In some countries, the legislation for radiation protection is being revised in order to be harmonized with the international basic safety standards of the International Atomic Energy Agency [5]. These new regulations require the application of most recent dose evaluation models, including the ICRP human respiratory tract model and the newly developed biokinetic models. However, the implementation of the new models in computers is complicated and software as of yet scarce. The present work is our contribution to es-

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tablishing a new model for estimating excretion rates following the intake of nine different radionuclides.

BIOKINETIC MODELS

In terms of the intake or committed effective dose, the knowledge of the behavior of radioactive materials within the human body is essential for the interpretation of measured activities in the body or excreta [9]. Biokinetic models are used to calculate body or organ content and daily urinary or faecal excretion at specified times after intake. Intakes of radionuclides can occur via a number of pathways. In occupational exposure, the main route of intake is by inhalation, although a fraction of any material deposited in the respiratory system will be transferred to the throat and swallowed, creating an opportunity for absorption into the gastrointestinal tract [10]. Intakes by direct ingestion may occur by absorption via intact skin.

In the case of occupationally exposed workers, the ICRP has developed models for describing the behavior of radionuclides that have entered the body either by inhalation or by ingestion [9]. Other possible pathways of exposure, intakes, are only likely to occur as a result of accidents that cannot be completely prevented by workplace control measures or readily predicted.

The biokinetic models developed by the ICRP are intended for use in ordinary circumstances. Evaluations of doses from measurements performed according to routine monitoring programmers are an example of this. The evaluation of doses in accidental situations requires more specific information about the time and pattern of intake, physiochemical forms of the radionuclides, and characteristics of the individual (*e. g.* body mass). Individual specific data on the biokinetic of a radionuclide may be obtained through special monitoring, *i. e.* by repeated direct measurements of the whole body or specific sites and measurements of excretion [11]. Details of the biokinetic models of the human respiratory tract for radiological protection purposes have been issued by the ICRP-66 [7].

MONDAL SOFTWARE

The personal computer based software Mondal used in this work provides a useful tool for dosimetrists involved in radiation protection for assessing intakes of radionuclides and the resulting tissue equivalent and effective doses from bioassay measurements for both workers and members of the public. Mondal software is distributed by the National Institute of Radiological Sciences (NIRS), Anagawa, Japan.

Basically, the software consists of a data library for fractions of inhaled or ingested radioactivity retained in the entire body or a specific organ, excreted daily into urine or faeces, hereafter referred to as the intake retention fraction (IRF).

If single acute intake is assumed, the activity $m(t)$ of intake I, is calculated simply from the IRF value at measurement day t and measured activity M. In case of a chronic intake for T days, the values of IRF at measurement day, t, are $m(T+t-1)$ for the intake on the first day, $m(T+t-2)$ for the intake on the second day and $m(T+t-i)$ for the intake on the ith day. Hence, in this program, an approximate value of intake is calculated from a set of called values of IRF, $m(T+t-1)$, $m(T+t-2)$, $m(T+t-3)$, …, $m(t)$, and the measured activity, M, by the equation

$$I = \frac{TM}{\sum_{i=1}^{T} m(T+t-i)} \quad (1)$$

If the intake differs from day to day, the measured activity can, approximately, be expressed in the form of

$$M = I_1 m(T+t-1) + I_2 m(T+t-2) + \ldots + I_i m(T+t-i) + \ldots + I_T m(t) \quad (2)$$

where I_i is the activity of intake at the ith day. If relative values of intake, H_i, are given, eq. 2 becomes

$$M = C \sum_{i=1}^{T} H_i m(T+t-i) \quad (3)$$

For a constant C, an approximate value of the total intake can, therefore, be expressed as

$$I = \frac{\sum_{i=1}^{T} H_i}{\sum_{I=1}^{T} H_i m(T+t-i)} M \quad (4)$$

By taking the working hours in each day as the relative values of the daily intake, an approximate value of the total intake is calculated from a set of values of IRF, $m(T+t-1)$, $m(T+t-2)$, $m(T+t-3)$, …, $m(t)$, and the measured activity, M using eq. 4.

In order to determine tissue equivalent doses and effective doses delivered for various periods after the intake, the activity of intake calculated above is multiplied by the dose conversion coefficient (S_v/B_q) given in the ICRP Database of Dose Coefficients [12].

CONSTRUCTED MODEL

The present work is a presentation of our simulation program for estimating excretion rates following the intake of nine different radionuclides: cobalt, iodine, cesium, strontium, ruthenium, radium, thorium, plutonium, and uranium, using a series of exponential functions representing the urinary and faecal excre-

tion. The clearance of inhaled material from the compartment is described by a set of interlinked first order differential equations that estimate the urinary and faecal excretion over a prolonged period [9, 10]. In addition to the assessment of the intake and doses of radionuclides, these differential equations are used to calculate the activity in the daily urinary or faecal excreta. The data provided cover periods of up to 1000 days.

Following the entry of cobalt into the blood, a large fraction is rapidly excreted [4], recommending a model in which 50% of cobalt reaching the circulation is rapidly excreted with a biological half-life of 0.5 days, where 5% is taken up by the liver and 45% uniformly distributed to all other tissues. Fractions of 0.6 and 0.2 are assumed to be lost from the liver and other tissues with a biological half-life of 6, 60, and 800 days, respectively. This model was then endorsed by ICRP-67 [13], which also recommended a value for the urinary to faecal excretion ratio of 6:1. Thus, urinary excretion can be given by

$$u(t) = 0.49 e^{-0.693\frac{t}{0.5}} + 2.4 \cdot 10^{-2} e^{-0.693\frac{t}{6}}$$
$$+ 8.1 \cdot 10^{-6} e^{-0.693\frac{t}{60}} + 6.1 \cdot 10^{-5} e^{-0.693\frac{t}{800}} \quad (5)$$

while the faecal excretion equals

$$f(t) = 0.21 e^{-0.693\frac{t}{0.5}} + 1.0 \cdot 10^{-2} e^{-0.693\frac{t}{6}}$$
$$+ 3.5 \cdot 10^{-4} e^{-0.693\frac{t}{60}} + 2.6 \cdot 10^{-5} e^{-0.693\frac{t}{800}} \quad (6)$$

The metabolic model used for iodine is based on that described by Rigg [14] and modified by ICRP. When iodine enters the transfer compartment, 0.3% is taken up by the thyroid with a half-life of 80 days. Iodine is lost from the thyroid in the form of organic iodine which is then assumed to be uniformly distributed among all organs and tissues of the body and retained with a biological half-life of 12 days. 90% of organic iodine is returned to the transfer compartment and the rest is excreted via faeces. Using Rigg's model, this leads to the following urinary excretion function

$$u(t) = 1.9 e^{-0.693\frac{t}{0.24}} + 1.9 \cdot 10^{-3} e^{-0.693\frac{t}{11}}$$
$$+ 1.9 \cdot 10^{-3} e^{-0.693\frac{t}{120}} \quad (7)$$

while faecal excretion is given by

$$f(t) = 5.1 \cdot 10^{-5} e^{-0.693\frac{t}{0.24}} + 2.6 \cdot 10^{-4} e^{-0.693\frac{t}{11}}$$
$$+ 2.6 \cdot 10^{-4} e^{-0.693\frac{t}{120}} \quad (8)$$

The retention in the thyroid is described by the following function

$$h(t) = 0.33 e^{-0.693\frac{t}{0.24}} - 1.8 \cdot 10^{-2} e^{-0.693\frac{t}{11}}$$
$$- 0.13 e^{-0.693\frac{t}{120}} \quad (9)$$

The model of cesium given in ICRP-30 [4] was also recommended in ICRP-56 [15] and used in ICRP-68 [16]. Cesium is distributed uniformly throughout all body tissues, 10% of activity is assumed to be retained with a biological half-life of 2 days (A) and 90% with that of 110 days (B). However, for females, the biological half-life for compartment B is significantly shorter than for males ICRP-56 [15]. There is also evidence that, in some countries, the mean biological half-life for cesium in adult males is shorter by about 110 days [17]. A urinary to faecal excretion ratio of 4:1 is recommended in ICRP-67 [13]. It is assumed that 80% of cesium lost from the body appears in urine. Thus, the following urinary excretion function is used

$$u(t) = 2.8 \cdot 10^{-2} e^{-0.693\frac{t}{2}} + 4.5 \cdot 10^{-3} e^{-0.693\frac{t}{110}} \quad (10)$$

and faecal excretion is given by

$$f(t) = 6.9 \cdot 10^{-3} e^{-0.693\frac{t}{2}} + 1.1 \cdot 10^{-3} e^{-0.693\frac{t}{110}} \quad (11)$$

The physiologically based recycling model for strontium is taken from ICRP-67 [13]. Alkaline earth elements strontium and radium follow the movement of calcium in the body, but exhibit different transfer rates from calcium due to the discrimination by biological membranes and bone minerals. Activity entering the blood (plasma) from the respiratory or gastrointestinal tract is retained by bone and soft tissues or excreted in the urine and faeces. All activities leaving the soft tissue compartments are assumed to be returned to the plasma. The activity returned to the plasma is assumed to be redistributed among tissues and excreta according to sane parameter values, as for the original input to plasma (ICRP-78) [9]. Ratios of urinary to faecal excretion, f_u/f_f, are variously given as 3~10 (ICRP-10) [18], and 2~6 [19]. A value of 4 has been adopted in our simulation program, $i. e. f_u = 0.8$ and $f_f = 0.2$. The urinary excretion is given by

$$u(t) = 0.13 e^{-0.693\frac{t}{3}} + 1.3 \cdot 10^{-3} e^{-0.693\frac{t}{44}}$$
$$+ 2.4 \cdot 10^{-5} e^{-0.693\frac{t}{4000}} \quad (12)$$

and faecal excretion is

$$f(t) = 3.4 \cdot 10^{-2} e^{-0.693\frac{t}{3}} + 3.2 \cdot 10^{-4} e^{-0.693\frac{t}{44}}$$
$$+ 5.9 \cdot 10^{-6} e^{-0.693\frac{t}{4000}} \quad (13)$$

For ruthenium absorbed by body fluids, data show that the subsequent tissue distribution is fairly

uniform. The ICRP-30 [4] recommended a model using three terms of retention expressions, 35% of activity is retained with a biological half-life of 8 days, 30% of 35 days, and 20% of a 1000 days. The biological half-life in the body is taken to be 0.3 days, from which 15% is assumed to be excreted directly. This model was later endorsed in ICRP-56 [15] and ICRP-67 [13] recommended that a urinary to faecal excretion ratio of 4:1 can be assumed for ruthenium. The urinary excretion is given as

$$u(t) = 0.28e^{-0.693\frac{t}{0.3}} + 2.4 \cdot 10^{-2}e^{-0.693\frac{t}{8}} + 4.8 \cdot 10^{-3}e^{-0.693\frac{t}{35}} + 1.1 \cdot 10^{-4}e^{-0.693\frac{t}{1000}} \quad (14)$$

and faecal excretion is

$$f(t) = 6.9 \cdot 10^{-2}e^{-0.693\frac{t}{0.3}} + 6.1 \cdot 10^{-3}e^{-0.693\frac{t}{8}} + 1.2 \cdot 10^{-3}e^{-0.693\frac{t}{35}} + 2.8 \cdot 10^{-5}e^{-0.693\frac{t}{1000}} \quad (15)$$

The physiologically based recycling model for radium is taken from ICRP-67 [13]. The model describes the kinetics of radium in bone, which is the main site of deposition and retention, and also considers retention in the liver and other soft tissues, as well as routes of excretion. It takes into account the initial intake into bone surfaces, transfer from surface to bone volume, and recycling from bone and other tissues to plasma. Measurements in individuals with radium burdens gave a mean faecal excretion of 95% and a mean urinary excretion of 5% [20]. The urinary excretion is given by

$$u(t) = 4.7 \cdot 10^{-2}e^{-0.693\frac{t}{0.4}} + 2.0 \cdot 10^{-3}e^{-0.693\frac{t}{5}} + 6.6 \cdot 10^{-5}e^{-0.693\frac{t}{60}} + 2.0 \cdot 10^{-6}e^{-0.693\frac{t}{700}} + 1.4 \cdot 10^{-7}e^{-0.693\frac{t}{5000}} \quad (16)$$

The faecal excretion is given as

$$f(t) = 0.89e^{-0.693\frac{t}{0.4}} + 3.9 \cdot 10^{-2}e^{-0.693\frac{t}{5}} + 1.3 \cdot 10^{-3}e^{-0.693\frac{t}{60}} + 3.8 \cdot 10^{-5}e^{-0.693\frac{t}{700}} + 2.7 \cdot 10^{-6}e^{-0.693\frac{t}{5000}} \quad (17)$$

For thorium absorbed in the blood, the main sites of deposition are the liver and skeleton. A generic actinide model was recommended in ICRP-67 [13]. The biological half-life of the transfer compartment is taken to be 0.5 day. For thorium entering the transfer compartment, 70% is assumed to be transferred to the bones where it is retained with a biological half-life of 700 days, while 16% is assumed to be uniformly distributed among all other organs and tissues of the body

where it is retained with a biological half-life of 7000 days. The remaining 10% of thorium entering the transfer compartment is assumed to go directly to excretion. The urinary excretion is

$$u(t) = 0.14e^{-0.693\frac{t}{0.5}} + 2.0 \cdot 10^{-4}e^{-0.693\frac{t}{700}} + 6.1 \cdot 10^{-5}e^{-0.693\frac{t}{8000}} \quad (18)$$

For plutonium absorbed in the blood, the main sites of deposition are the liver and skeleton. The model of plutonium is given in ICRP-67 [13]. When plutonium enters the transfer compartment, 45% is assumed to be transferred to the liver and 45% to the bones (ICRP-18) [21]. Retention half-life in liver is taken to be 20 years and 50 years in the bones (ICRP-48) [22]. The fraction transferred to the gonads is $3.5 \cdot 10^{-4}$ for males and $1.1 \cdot 10^{-4}$ for females. Most excretion functions are based on recent data. These data, together with experimental animal data, were used to produce a series of exponential functions to represent urinary and faecal excretion. The urinary excretion is given by

$$u(t) = 4.1 \cdot 10^{-3}e^{-0.693\frac{t}{1.2}} + 1.2 \cdot 10^{-3}e^{-0.693\frac{t}{5.5}} + 1.3 \cdot 10^{-4}e^{-0.693\frac{t}{42}} + 3.0 \cdot 10^{-5}e^{-0.693\frac{t}{300}} + 1.2 \cdot 10^{-5}e^{-0.693\frac{t}{4000}} \quad (19)$$

and faecal excretion equals

$$f(t) = 6.0 \cdot 10^{-3}e^{-0.693\frac{t}{2.0}} + 1.6 \cdot 10^{-3}e^{-0.693\frac{t}{6.6}} + 1.2 \cdot 10^{-4}e^{-0.693\frac{t}{56}} + 2.0 \cdot 10^{-5}e^{-0.693\frac{t}{3800}} + 1.2 \cdot 10^{-5}e^{-0.693\frac{t}{4000}} \quad (20)$$

The physiologically based recycling model for uranium is recommended in ICRP-69 [23]. It describes in detail the kinetics of uranium in bone and also considers retention in the liver, kidneys, and other soft tissues, as well as routes of excretion. Of the uranium entering the transfer compartment, 54% is directly excreted, fraction of 20% is transferred to bone minerals and 2.3% is retained there with a biological half-life of 20-5000 days. A fraction of 12% is assumed to be transferred to the kidneys and 0.052% is retained there with a biological half-life of 6-1500 days. A fraction of 12% is assumed to go to all other tissues of the body and 0.032% retained there with a biological half-life of 6-1500 days. Thus, the urinary excretion equals

$$u(t) = 1.5e^{-0.693\frac{t}{0.25}} + 2.8 \cdot 10^{-2}e^{-0.693\frac{t}{6}} + 6.9 \cdot 10^{-3}e^{-0.693\frac{t}{20}} + 4.8 \cdot 10^{-7}e^{-0.693\frac{t}{1500}} + 2.7 \cdot 10^{-8}e^{-0.693\frac{t}{5000}} \quad (21)$$

RESULTS AND DISCUSSION

Figure 1 represents the daily faecal excretion rates estimated by both Mondal (closed circles) and our constructed model (open circles), after the intake of radionuclides cesium (top), strontium (middle), and ruthenium (bottom).

Figure 2 shows the daily faecal excretion rates estimated by both Mondal (closed circles) and our constructed model (open circles) after the intake of radionuclides.

The curves from top to bottom correspond to the radionuclides under investigation: radium, plutonium, cobalt, and iodine, respectively. It has been shown that: strontium provided the best fitting with the corre-
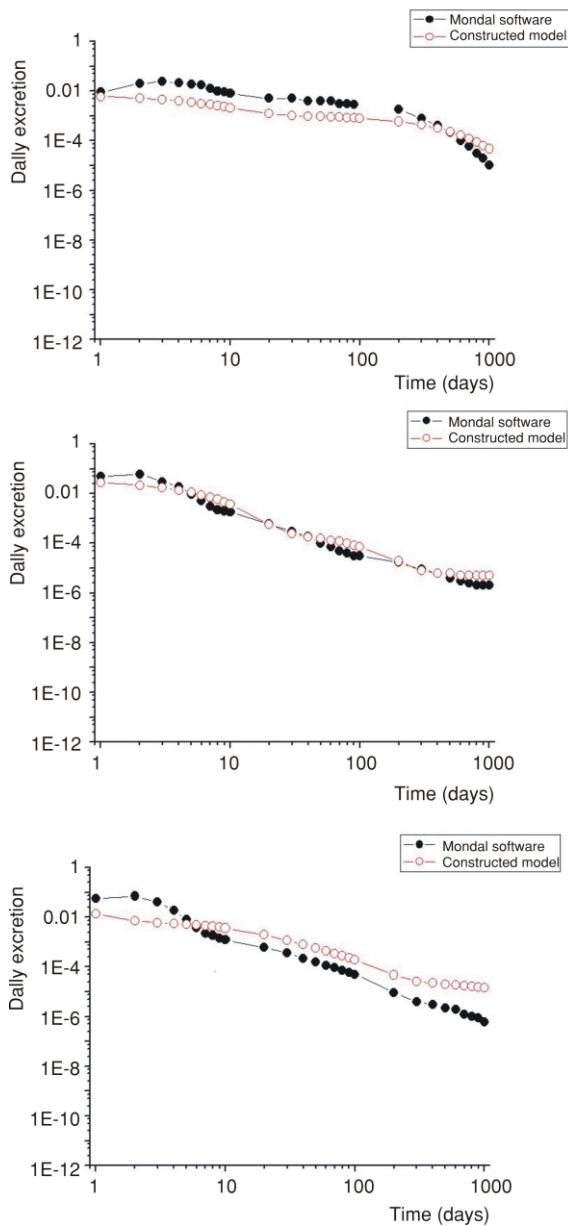

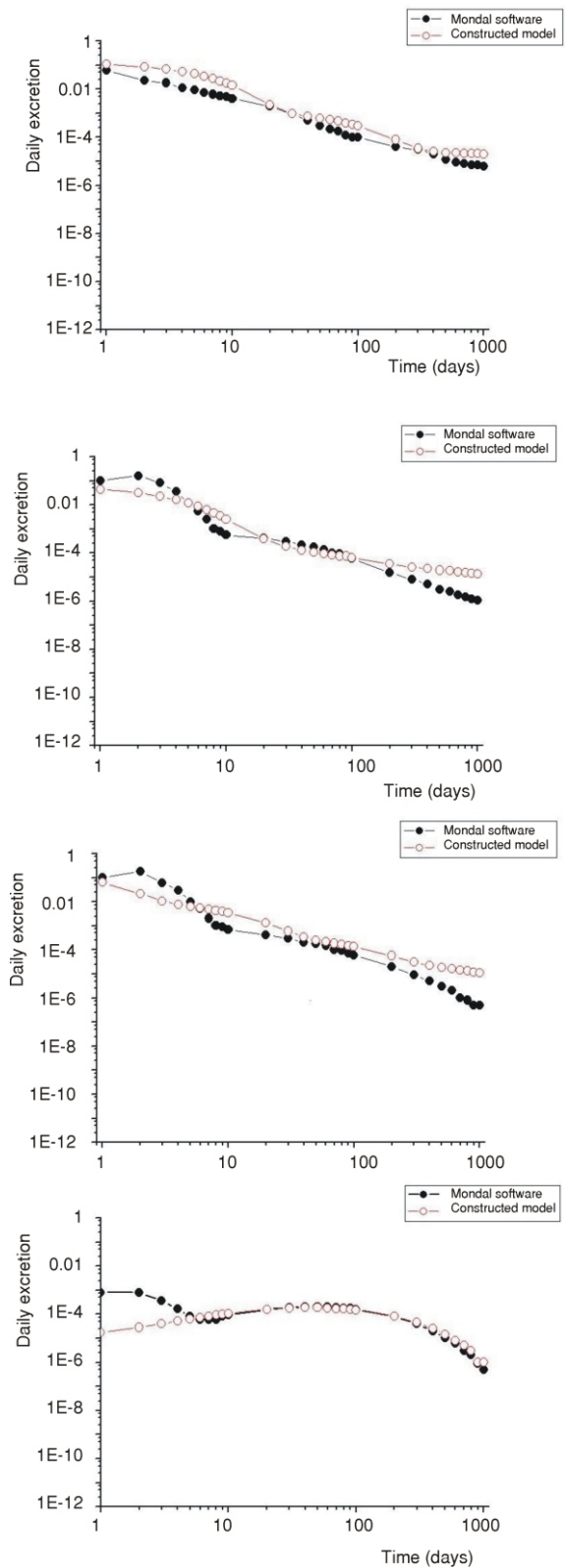**Figure 1. Faecal excretion rates after the intake of cesium, strontium, and ruthenium, respectively**

Figure 2. Faecal excretion rates after the intake of radium, plutonium, cobalt, and iodine, respectively

lation coefficient R^2 equal ($R^2 = 0.85$), followed by plutonium ($R^2 = 0.81$), cesium ($R^2 = 0.77$), ruthenium ($R^2 = 0.62$), radium ($R^2 = 0.53$), cobalt ($R^2 = 0.51$), and iodine ($R^2 = 0.35$), respectively.

Figure 3 shows the daily urinary excretion rates estimated by both Mondal and our constructed model after the intake of the radionuclides. The curves from top to bottom correspond to radionuclides cesium, strontium, and ruthenium, respectively.

Figure 4 represents daily urinary excretion rates estimated by both Mondal and our constructed model after the intake of radionuclides. The curves from top to bottom correspond to the radionuclides: radium, plutonium, and cobalt, respectively.
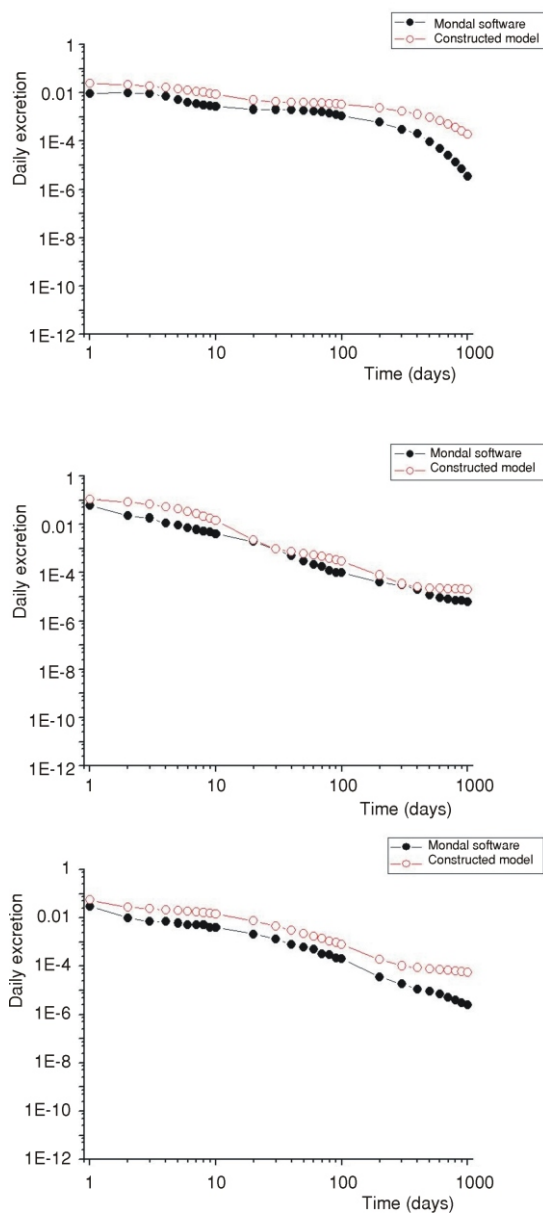

Figure 3. Urinary excretion rates after the intake of cesium, strontium, and ruthenium, respectively
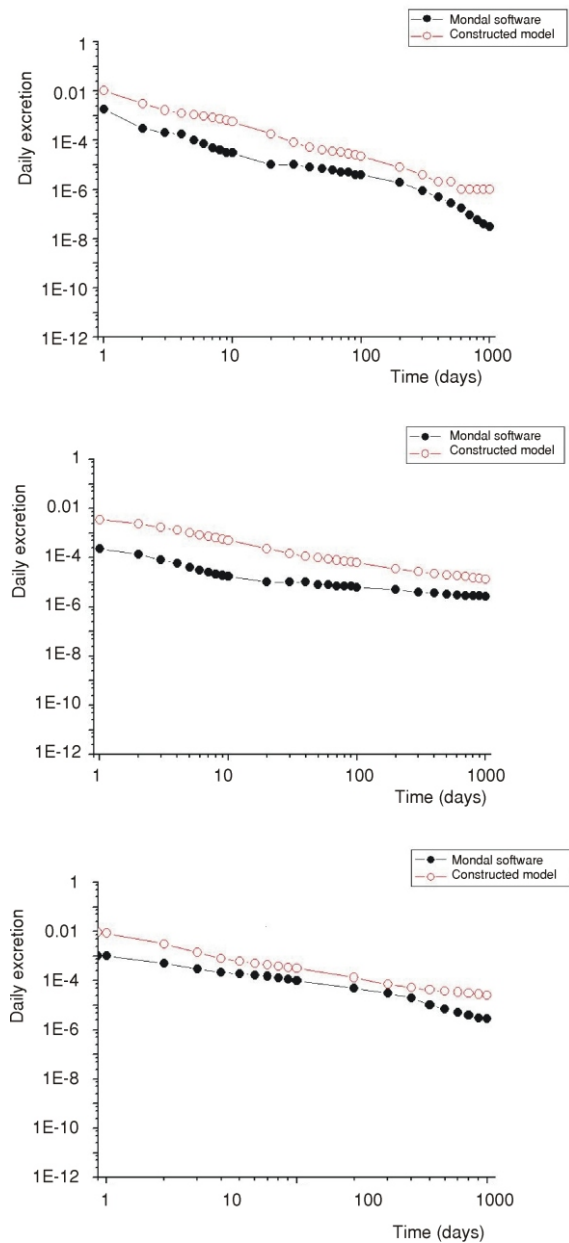

Figure 4. Urinary excretion rates after the intake of radium, plutonium, and cobalt, respectively

Figure 5 shows the daily urinary excretion rates estimated by both Mondal and our constructed model after the intake of radionuclides. The curves from top to bottom correspond to the radionuclides: iodine, thorium, and uranium, respectively. It has been shown that iodine provided the best fit ($R^2 = 0.99$), followed by cobalt ($R^2 = 0.98$), radium ($R^2 = 0.97$), thorium ($R^2 = 0.96$), cesium ($R^2 = 0.95$), plutonium ($R^2 = 0.95$), ruthenium ($R^2 = 0.91$), uranium ($R^2 = 0.86$), and strontium ($R^2 = 0.81$).
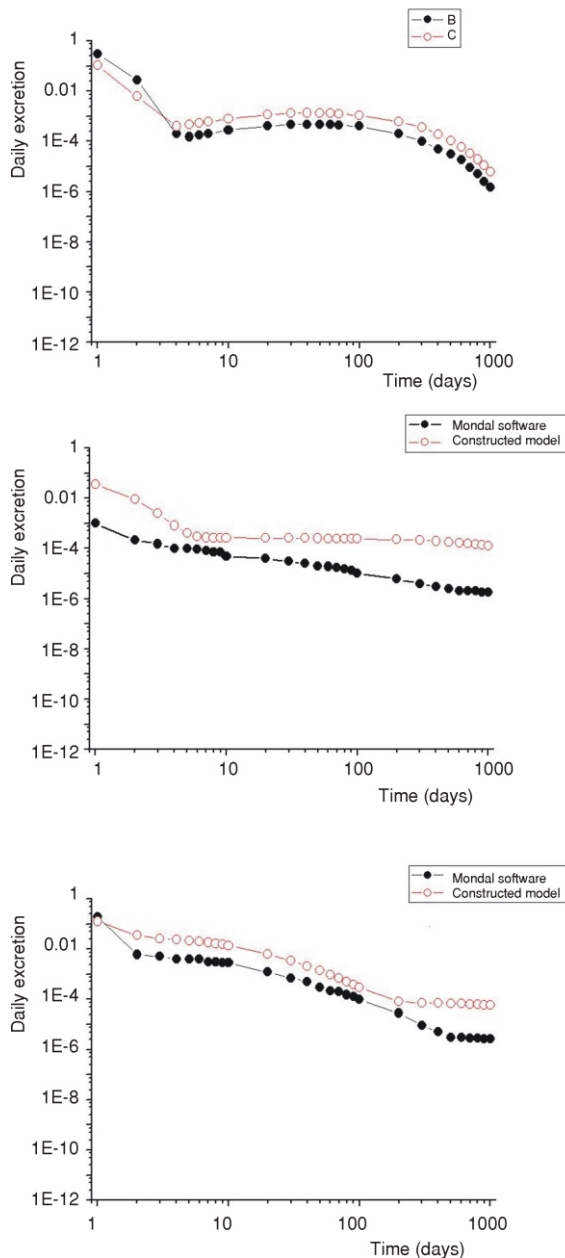
Figure 5. Urinary excretion rates after the intake of iodine, thorium, and uranium, respectively

CONCLUSIONS

The committed effective dose has been calculated by our model to obtain the urinary and faecal excretion rates for each radionuclide. The constructed model is further validated by a comparison with the widely spread Mondal simulation program. The results of the daily faecal excretion rates estimated by both Mondal and our constructed model after the intake of radionuclides under investigation show that strontium provided the highest fit with ($R^2 = 0.85$) and iodine the lowest one with ($R^2 = 0.35$).

The results of the daily urinary excretion rates estimated by both Mondal and our constructed model after the intake of radionuclides under investigation show that iodine provided the highest fit with ($R^2 = 0.99$), with strontium showing the lowest fit with ($R^2 = 0.81$). As expected and mentioned in ICRP-78 [9] and ICRP-54 [3], the values of urinary excretion are higher than those of faecal excretion.

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НОВИ СИМУЛАЦИОНИ МОДЕЛ ЗА ПРОРАЧУН ИНТЕРНОГ ИЗЛАГАЊА НЕКИМ НУКЛИДИМА

У раду је приказан нови модел за процену брзине излучивања која следи по уношењу девет различитих радионуклида, а заснива се на редовима математичких функција. Размотрани су кобалт, јод, цезијум, стронцијум, рутенијум, радијум, торијум, плутонијум и уранијум. Моделом су одређене ефективне дозе и добијене брзине уринарног и фекалног излучивања за сваки радионуклид. Модел је потом потврђен поређењем са резултатима опште прихваћеног Мондал софтвера и једног симулационог програма. Добијени резултати показују сагласност Мондал програмског пакета и изграђеног модела.

Кључне речи: радионуклиди, Мондал програмски пакет, симулација