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Determination of Uranium Internal Dose Exposure through Soil Digestion Using RDRC and URODC Software

(Penentuan Dos Dedahan Dalaman Uranium Melalui Penghadaman Tanih Menggunakan Perisian RDRC dan URODC)

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ABSTRACT

This study was conducted to determine the dose of internal exposure through ingestion of soil in the vicinity of the repository facility in Bukit Kledang, Ipoh, Perak. Data from this study can assess the risk of radiation exposure to the health of local population, specifically blood, liver and bone cancers. Activities of radionuclide ^{238}U in MG and M10 in the gastric phase are 1.118 ± 0.062 and 1.232 ± 0.073 Bq/kg, while the respective activities in the gastrointestinal phase are 0.553 ± 0.051 and 0.905 ± 0.082 Bq/kg. Samples of M10 recorded the highest reading of internal exposure in both phases. Digestion of 2 g soil from M10 samples on gastric phase generated the annual effective dose of 3.168 µSv/year with an assessment of cancer risk by 0.001% within 70 years to public. Organ dose for blood, liver and bone were 0.59, 11.60 and 65.95 µSv, respectively. Analysis of organ doses based on the concentration of ^{238}U found that M10 has higher dose compared to MG. Risk assessment predicted for 70 years after the ingestion of the soil for blood cancer was 0.003% and liver cancer was 0.004% while the highest cancer risk was for bone cancer with 0.023%. Although the concentration of specific activity of ^{238}U identified is low, it is shown that the internal dose exposure as a result of digestion of radionuclides are below the standard and can be considered as safe for public.

Keywords: Cancer risk; internal dose exposure; repository facility; soil sample; ^{238}U

ABSTRAK

Kajian ini dijalankan untuk menentukan dos dedahan dalaman melalui penghadaman tanih di kawasan sekitar fasiliti repositori di Bukit Kledang, Ipoh, Perak. Data daripada kajian ini dapat menilai risiko dedahan sinaran terhadap kesihatan penduduk setempat khususnya penyakit kanser darah, hati dan tulang. Aktiviti spesifik radionuklid ^{238}U di MG dan M10 pada fasa gastrik masing-masing adalah 1.118 ± 0.062 dan 1.232 ± 0.073 Bq/kg, manakala nilai aktiviti spesifik tanih di MG dan M10 pada fasa gastrousus pula masing-masing adalah 0.553 ± 0.051 dan 0.905 ± 0.082 Bq/kg. Sampel M10 merekodkan bacaan dos dedahan dalaman tertinggi berbanding dengan sampel MG pada kedua-dua fasa. Penghadaman 2 g tanih pada fasa gastrik sampel M10, merekodkan dos berkesan tahunan sebanyak 3.168 µSv/tahun dengan penilaian risiko kanser sebanyak 0.001% dalam jangka masa 70 tahun untuk orang awam. Dos organ bagi darah, hati dan tulang masing-masing adalah sebanyak 0.59, 11.60 dan 65.95 µSv. Dos organ ini juga dianalisis berdasarkan kepekatan ^{238}U. Didapati bahawa dos ini lebih tinggi untuk M10 berbanding MG. Penilaian risiko dalam jangka masa 70 tahun bagi kanser darah adalah 0.003%, kanser hati sebanyak 0.004% manakala paling tinggi untuk mendapat kanser tulang iaitu sebanyak 0.023%. Lantaran nilai kepekatan aktiviti ^{238}U yang dikenal pasti adalah rendah maka ia membuktikan bahawa dedahan sinaran dan dos dedahan dalaman sinaran akibat penghadaman radionuklid ini adalah di bawah piawai yang ditetapkan.

Kata kunci: Dos dedahan dalaman; fasiliti repositori; risiko kanser; sampel tanih; ^{238}U

INTRODUCTION

Recently, the Long Term Storage Facility (LTSF) in Bukit Kledang, Perak, Malaysia, has been upgraded to repository facility upon the completion of decontamination and decommissioning (D&D) process. It is a radioactive waste disposal site where waste of rare earth processing is mostly thorium produced by itirium extraction from monazite (AELB 2016). But now, after decades, the disposal site still needs to be monitored for its radioactivity to ensure it is safe for the surrounding population.

It is common to know that some of the chemical elements present in the environment are naturally radioactive. One of them is uranium that is always present in the crust and in the tissues of all living beings. Natural radioactivity arises mainly from the primordial radionuclides, such as ^{40}K, and the radionuclides from ^{238}U and ^{232}Th series and their decay products, which are present at trace levels in all ground formations (Tzortzis et al. 2004). Natural uranium consists of three isotopes where 99.27% of them are ^{238}U (Yasmin et al. 2016). The behaviour of natural radioactivity is important to be studied

as it is useful for radiological assessment (Al Kharouf et al. 2008). Exposure to ^{238}U contaminants via ingestion of soil is routinely estimated as part of a risk assessment to the contaminated sites. Unintentional consumption of contaminants may also happen as a result of poor personal hygiene and unwashed vegetables (Intawongse & Dean 2006). Ingestion of ^{238}U radionuclide could lead to chemical and radiological toxicity to organs such as bone, kidney and blood (ATSDR 2013).

Based on previous studies, the radiation exposure dose by uranium in this soil can be done by obtaining the concentration of the radionuclide content first, using various types of gamma-ray detection systems such as ICP-MS (Hollriegl et al. 2010; Traber et al. 2015), Neutron Activation Analysis (NAA) (Daniela & Kratz 1996) and X-ray Fluorences analysis (XRF) (Yasmin et al. 2016). The *in vitro* digestive method (DIN method) is a method that has been identified as one of the mimic and similar methods of human digestive system in the gastric and intestinal stage. This method introduced by Traber et al. (2015) is a DIN method that is a combination of soil with intestinal and gastric juice as in the digestive system (gastrointestinal tract). Many studies have been done using the DIN method, among which are recent studies to determine the concentration of uranium intake through soil digestion (Yasmin et al. 2016). By knowing this uranium concentration, internal doses can be determined using various methods of analysis whether using specific software such as SAAM II or computation using a specific dose equation and biocinetic model (Traber et al. 2015).

The aim for this study was to determine the dose of internal exposure through ingestion of soil in the vicinity of the disposal of radioactive waste by using Radiation Dose to Risk Converter (RDRC) and Uranium Organ Radiation Dose Calculator (URODC). The data that has been analyzed will be used to estimate the risk of cancer as a result of this exposure to the human digestive system. In addition, this study helps identify the internal level of exposure received by the public and the safety of soil-sourced food in the area.

Materials and Methods

Sampling Area

This study has been carried out in the repository facility, Bukit Kledang, Perak, which has been upgraded from The Long Term Storage Facility (LTSF) after the completion of decontamination and decommissioning (D&D) process. This area is the radioactive waste storage site for the Asian Rare Earth Factory (ARE). The sampling station is located in the vicinity of the facility which is also close to the water source of the Sungai Johan. The sampling location shown in Figure 1.

Samples Collection and Preparation

In this study, dose monitoring was carried out at the sampling stations by using survey meter with Geiger Muller (GM) detector (μSv/hr) and Sodium Iodide (NaI) detector to detect alpha, beta and gamma. The measurement had been done according to a specified distance of 5 cm and 1 m above the ground. All the samples were collected at a depth of 0-10 cm from the soil-surface using hand Auger and were carried in dried cleaned polyethylene bags with sample codes and transferred to the laboratory. The soil samples were cleaned and dried in the oven at 75°C for 3 days. Samples were pulverized and sieved (500 micron) to ensure that the sample is homogeneous.

Gastrointestinal *In Vitro* Digestive Method (DIN Method)

The assessment and measurement of uranium radiation exposure doses are calculated through radionuclide concentration in the soil. Among the methods that have

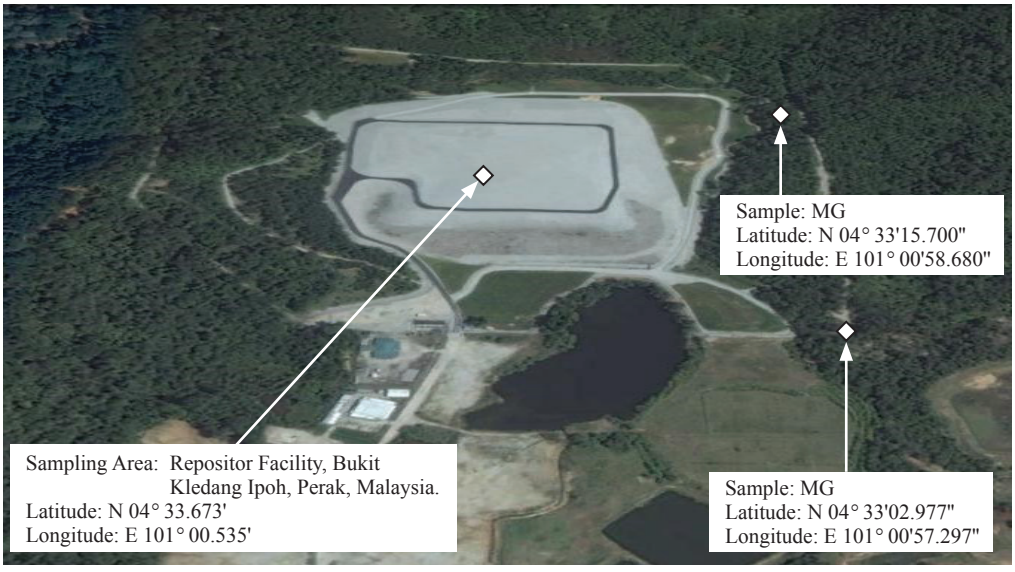

FIGURE 1. Location of sampling station in the repository facility

been identified to determine the dose is the accumulation of soil in the human body, through the digestive process. Introduced by Traber et al. (2015), the *in vitro* digestive method (DIN method) is a combination of soil with intestinal and gastric fluid as it exists in the digestive system (gastrointestinal tract). DIN method is a method that has been identified as one of the mimic and similar methods of human digestive system in the gastric and intestinal stage. Table 1 shows the preparation of gastric fluid and gastrointestinal fluid. According to Höllriegl et al. (2010), it is important to ensure that the gastric fluid and intestinal fluid are used to conform or completely mimic the human digestive system. Figure 2 shows the sequence of DIN method for this study.

Briefly, 2 g of soil (duplicated) was incubated at pH 2.0 with 100 mL of artificial gastric fluid for 2 h. This was followed by the addition of 100 mL of artificial gastrointestinal fluid at pH 7.5 for 6 h. Temperature was maintained at 37°C in a water bath and agitation performed at 200 rpm using a stirrer throughout incubation process. After 2 h, two aliquots of each 10 mL were drawn from gastric soil solution, centrifuged at 3500 rpm for 15 min and filtrated. Remaining gastric solution was mixed with solid bicarbonate and 100 mL of intestinal fluid to form a mixture of gastrointestinal soil solution. After incubated for 6 h with a soil-to-fluid ratio of 1:25, two aliquots of each 10 mL were withdrawn, centrifuged and filtrated.

TABLE 1. Preparation for gastric and intestinal fluids

DIN method		
	Gastric fluid	Gastrointestinal fluid
Sodium chloride (NaCl)	2.9	-
Pottasium cloride (KCl)	0.7	0.3
Pottasium phosphate (KH_2PO_4)	0.27	-
Calcium cloride ($CaCl_2 \times 2\ H_2O$)	-	0.5
Magnesium cloride ($MgCl_2 \times 6\ H_2O$)	-	0.2
Sodium bicarbonate ($NaHCO_3$)	-	1
Pepsin	1	-
Mucin	3	-
Tripsin	-	0.3
Pankreatin	-	9
Bile bovine	-	9
Urea	-	0.3
Hydrocloric acid 30% (HCl)	pH adjustment	
Sodium bicarbonate $NaHCO_3$		pH adjustment
Final pH	2.0	7.5

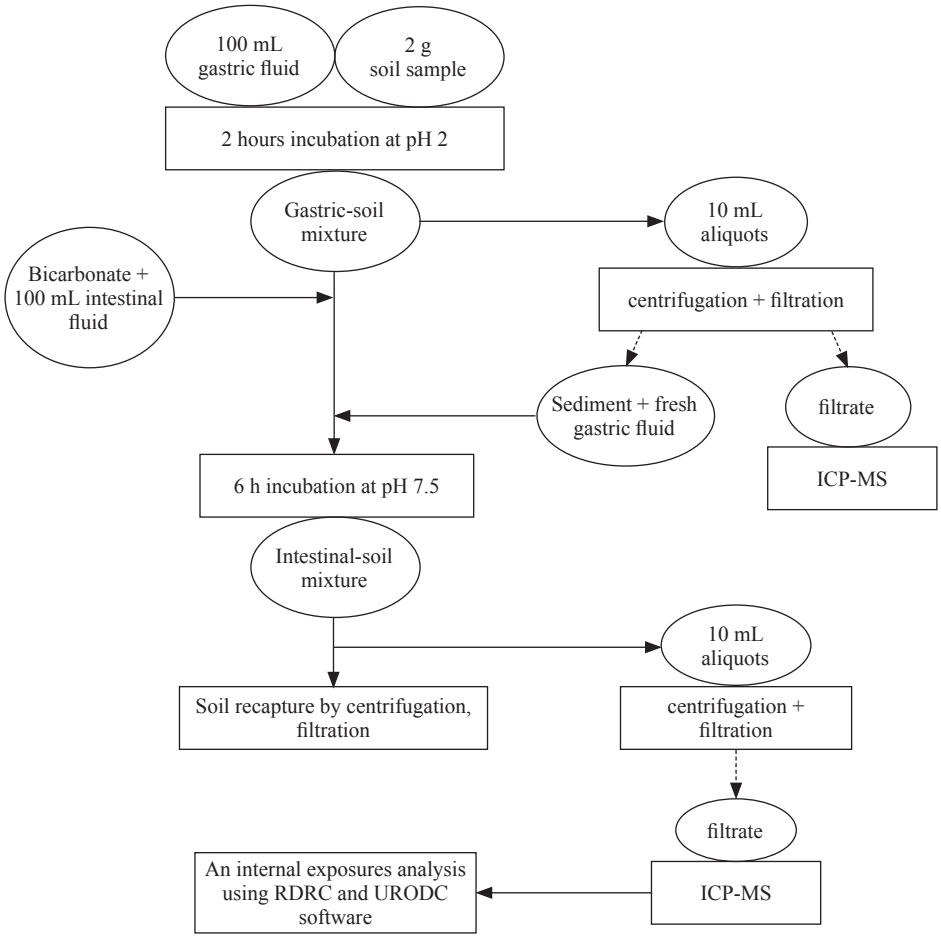

FIGURE 2. The sequence of DIN method

The solutions were stored frozen until measured with inductively coupled plasma mass spectrometer (ICP-MS) (Hollriegl et al. 2010; Traber et al. 2015).

CONCENTRATION DETERMINATION

The concentration of ^{238}U radionuclide was calculated based on (1):

$$\rho(\text{mg/kg}) = \frac{c(\mu g/L) \times V(\text{L})}{M(g)} \quad (1)$$

where ρ is the sample concentration (mg/kg); V is the volume of the sample (L); c is the mean of the sample from the ICP-MS analysis ($\mu g/L$); and M is the mass of the sample (g).

This concentration should be converted into Bq/kg unit as a specific activity of ^{238}U radionuclide in the sample, using a ^{238}U conversion factor of 1 Bg/kg ^{238}U equivalent to 0.081 mg/kg (Qing 2011). This concentration value will be used in (2) to determine the annual effective dose.

CALCULATION OF EXPOSURE DOSAGE USING THE DOSE CALCULATION SOFTWARE METHOD

The concentration values from (1) will be used to determine the annual effective dose by using Radiation Dose to Risk Converter (RDRC) and Uranium Radiation Organ Dose Calculator (URODC) software. The concentration values are analyzed by this software and their internal exposure doses will be identified. Then the assessment of annual exposure dose, cancer risk, effective dose and dose on each organ in the digestive system can be calculated and analyzed.

$$D(\text{Sv/year}) = c(\text{Bq/kg}) \times P_t(\text{kg/year}) \times P_D(Sv/Bq) \quad (2)$$

where D is the annual effective dose (Sv/year); P_t is the annual intake (kg/year); c is the sample concentration (Bq/kg); and P_D is the dose coefficient (Sv/Bq).

The annual effective dosage (D) is calculated based on sample concentration (specific activity) multiplied by annual intake (soil) and dose coefficients (Pulhani et al. 2005). Annual effective dosage after 2 g of digestion will be determined using RDRC and URODC software based on (2) above.

$$R_k = D(\text{Sv/year}) \times J_h(\text{year}) \times R_f(^{1}\text{-Sv}) \quad (3)$$

where R_k is the life time cancer risk; J_h is the life expectancy (year); D is the annual effective dose (Sv/year); and R_f is the risk factor (^{1}-Sv).

The value of life time cancer risk (R_k) can be derived from the annual effective dose multiplication, life expectancy and risk factors (3). According to ICRP 72, life expectancy for the public is 70 years and risk factor is 0.05/Sv. This study uses the values recommended by ICRP 72 as used by URODC software.

Results and Discussion

THE CONCENTRATION OF ^{238}U IN SOIL SAMPLES AFTER DIGESTION PROCESS

Table 2 shows the concentration of ^{238}U content found in the soil from the sampling location around the repository facility, involving two phases in the digestive process i.e. the gastric phase and gastrointestinal phase. The concentration of ^{238}U was between 0.045 and 0.100 mg/kg lower than those reported by WHO (2003) and ATSDR (1999) as stated by WISE (2016). The level of ^{238}U radioactivity is calculated using the conversion rate for uranium i.e. 1 Bg/kg ^{238}U equivalent to 0.081 mg/kg (Qing 2011).

In the gastric phase, the ^{238}U radionuclide concentration for the M10 sample was 0.100 ± 0.006 mg/kg equivalent to 1.232 ± 0.073 Bq/kg, while for the MG sample was 0.091 ± 0.005 mg/kg equivalent to 1.118 ± 0.062 Bq/kg. Furthermore, in the gastrointestinal phase, the ^{238}U radionuclide concentration for the M10 sample was 0.073 ± 0.007 mg/kg equivalent to 0.905 ± 0.082 Bq/kg, while for the MG sample was 0.045 ± 0.004 mg/kg equivalent to 0.553 ± 0.051 Bq/kg.

The results showed that the concentration of ^{238}U in soil samples in M10 was higher than those in MG in the gastric and gastric phase. This may be related to the area's geographical factors. There is a possibility that it is related to the type of rock from which the soil originates and this was agreed upon from previous study (Lee & Wagiran 2014). The abundance of ^{238}U in M10 i.e. in downstream areas is likely to be higher than that of the upstream area including the MG. The abundance of this radionuclide is naturally in view of the fact that this area is an unexplored and untouched area.

Based on Figure 3, it is found that in the gastric phase, the specific activity concentration of ^{238}U soil in both samples is high compared with the gastrointestinal phase. The trend of difference in concentration of radionuclide activity in the gastric and gastric phase was also found to be similar to the previous studies (Table 3) conducted using *in vitro* digestive methods that mimic the digestive process using gastric juice and synthetic gastrointestinal juice (Höllriegl et al. 2010; Nur et al.

TABLE 2. The concentration and specific activity of ^{238}U in the soil through the digestive process

Sample	Radionuclide ^{238}U			
	Concentration (mg/kg)		Specific activity (Bq/kg)	
	Gastric phase	Gastro-intestinal phase	Gastric phase	Gastro-intestinal phase
M10	0.100 ± 0.006	0.073 ± 0.007	1.232 ± 0.073	0.905 ± 0.082
MG	0.091 ± 0.005	0.045 ± 0.004	1.118 ± 0.062	0.553 ± 0.051

TABLE 3. The average comparison of 238U radionuclide concentration in this study and previous studies

Reference	Studies area	Concentration (mg/kg)	Specific activity (Bq/kg)
This study (2017)	Repository Fasility, Perak	0.045 - 0.100	0.553 - 1.232
Yasmin et al. (2016)	Pahang, N. Sembilan, Selangor	0.018 - 0.001	0.006 - 0.603
WHO (2003)	Purata Dunia	0.006×10^{-1}/day 0.730/year	-
ATSDR (1999)	Amerika Syarikat	0.002/day 0.219/year	-

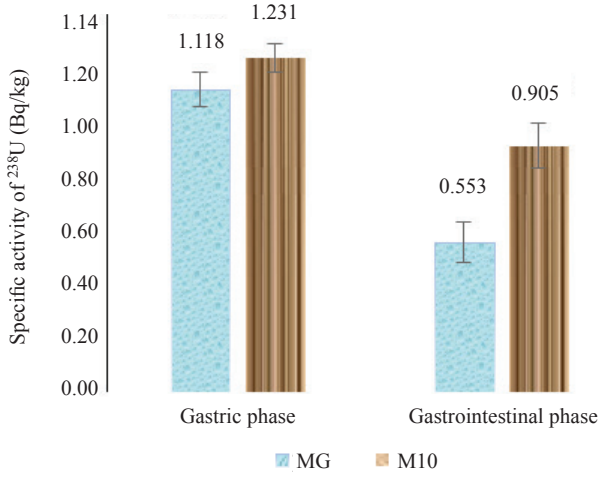

FIGURE 3. The concentration of specific activity of radionuclide ^{238}U soil samples in MG and M10 in digestive phase

2015; Yasmin et al. 2016). All of these studies relate the value of the pH difference in each phase affecting the value of radionuclide concentration ^{238}U in the digestive system. The difference in pH value in the gastric phase (pH 2) and in the gastrointestinal phase (pH 7), influenced the decomposition of ^{238}U in the digestive process to be absorbed into the human body (Höllriegl et al. 2010). The tendency of changing the pH value in the gastric phase to the gastrointestinal phase alters the acidic medium to alkali (Oliver 1999). This causes the ^{238}U radionuclide easier to dissolve in a more acidic state through chemical reactions (Nur et al. 2015).

INTERNAL EXPOSURE DOSE THROUGH SOIL DIGESTION

The annual effective dose rate through 2 g of soil digestion was analyzed using RDRD and URODC software, taking the specific concentration of ^{238}U activity in the soil through two digestive phases namely the gastric and gastrointestinal phase. Table 4 shows the annual effective dose obtained through the ^{238}U soil digestion assumed by 2 g per year.

The annual effective dose of M10 was 3.168 μSv/year in the gastric phase and 2.327 μSv/year in the gastrointestinal phase, while for MG it was 2.876 μSv/year in the gastric phase and 1.422 μSv/year in the gastrointestinal phase. This proves that the highest

TABLE 4. Annual effective dose found in each sample after 2 g of digestion per year

Sample	Phase	Annual effective dose (μSv/year)
MG	Gastric	2.876
	Gastrointestinal	1.422
M10	Gastric	3.168
	Gastrointestinal	2.327
UNSCEAR 2008		290

concentrations of ^{238}U radionuclide activity will give the highest effective annual dose and vice versa. The annual effective dose value in this study is between 1.422 and 3.168 μSv/year. This value is very low compared to the international standard values as reported in UNSCEAR (2008), where the average dose of natural radionuclide digest throughout the world for the public is estimated at 290 μSv/year.

CANCER RISK ASSESSMENT

Cancer risk assessment is performed using the highest annual effective dose rate through digestion of 2 g soil samples after 70 years according to ICRP 72 for the public (WISE 2016). The RDRC and URODC software are used to assess the risk of cancer by taking the highest concentration of specific activity content of ^{238}U, i.e. in M10 soil samples at the gastric phase of 1.232 Bq/kg. This value is taken into account for the calculation of dose rates based on assumptions that the estimate of the most high risk internal doses is at the concentration value of the specific activity concerned.

Table 5 shows a lifetime cancer risk assessment of any type of death cancer estimated for the public after digestion of 2 g of soil samples after 70 years. It was found that cancer risk assessment for fatal cancer are between 4.98×10^{-6} and 1.11×10^{-5} equivalents to 5 out of 1 000 000 people up to 1 out of 100,000 people can die of cancer after 70 years of digestion MG and M10. The highest risk of cancer was recorded based on the highest effective dose rate of 3.168 μSv/year (gastric phase for the M10) was 1.11×10^{-5} equivalent to 1 out of 100,000 people who had fatal cancer. This value was lower than reported by UNSCEAR (2000) i.e. 1.0×10^{-3} about 1 out

TABLE 5. Assessment of cancer risk for cancerous cancer for each sample in the digestive phase

Sample	Phase	Life time cancer risk	Life time cancer risk (%)
MG	Gastric	1.01×10^{-5}	1.01×10^{-3}
	Gastrointestinal	4.98×10^{-6}	4.98×10^{-4}
M10	Gastric	1.11×10^{-5}	1.11×10^{-3}
	Gastrointestinal	8.15×10^{-6}	8.15×10^{-4}
UNSCEAR 2000		1.00×10^{-3}	1.00×10^{-1}

** Cancer risk assessment estimated after 70 years (ICRP 72)

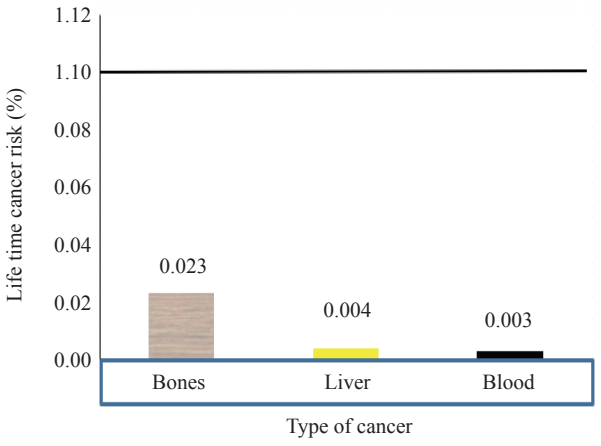

FIGURE 4. Cancer risk assessment based on the type of cancer (bones, liver and blood) analyzed by using URODC software

of 1000 people. It is clear here that digestion of 2 g of soil in the study area can be considered safe as it poses a low risk of cancer compared to those reported by UNSCEAR (2000).

However, after analyzing using URODC software, it was found that there were three organs that gave the highest dose value after digestion of 2 g soils of the study area i.e. bone, liver and blood organs. The organ dose obtained in this study was based on the highest concentration of ^{238}U radionuclide concentration in the gastrointestinal phase of M10 of 0.100 mg/kg. This value is used relative to other concentration values because it is considered as the concentration value which gives maximum risk to humans.

Referring to Table 6, it is found that the highest organ doses are in the bones of 65.95 μSv, followed by liver and blood organs of 11.6 and 8.41 μSv, respectively. The risk assessment of cancer according to the type of cancer in this study is calculated based on the dose in the organ and analyzed by software after 70 years of soil digestion of the study area. On the other hand, the risk of bone cancer based on organ doses is 2.31×10^{-4} or 0.023% equivalent to 2 out of 10 000 deaths with cancer of this type. The probability of getting liver cancer is 4.06×10^{-5} or 0.004% alone with 4 out of 100 000 death due to liver cancer. The risk of blood cancer are the least, where the probability of getting blood cancer is 2.94×10^{-5} or 0.003%, which is about 3 out of 100 000 deaths of cancer. The absorption of uranium into the human body starts from the digestive tract to the circulatory system which mostly occurs through the small intestine. According to Kathern and Burklin (2008), after the absorption of uranium from the intestine into the blood, it accumulates in the human body tissues especially on the bones and kidneys; which is the most sensitive target organ for uranium toxicity. Obviously here, that the dose of the organ in the bones is supposed to be the highest since bones are sensitive organ for uranium and this coincides with the results of this study.

Figure 4 shows all cancer risk assessment by type of cancer is below the reported value of UNSCEAR (2000) which is 1.0×10^{-3} equals 0.1% or 1 out of 1000 people suffering from cancers. Obviously, this study has shown that digestion of 2 g of soil in the study area after 70 years for the public is safe and below the standards set according to the assessment of low cancer risk. However, if the soil digestion persists and exceeds 2 g per year, the assessment of soil digestion should be carried out continuously to monitor the internal dose of its exposures so as not to exceed the prescribed limits.

CONCLUSION

In this study, concentration of specific activity of radionuclide ^{238}U in soil was measured based on 2 g of digestion per year. The concentration of soil sample activity in M10 was higher than the soil samples in MG in the gastric and gastrointestinal phase. This proves that the pH value affects the solubility of ^{238}U and also the geographical factor of the area plays a role in influencing the abundance of ^{238}U radionuclide in the soil. However, the value of this radionuclide concentration is considered to be low and below the value reported by WHO (2003) and ATSDR (1999). ^{238}U concentrations in both soil samples were analyzed using RDRC and URODC software and the highest annual effective dose was 3.168 μSv/year recorded in the gastrointestinal phase of M10. However, the dose value is also considered low as it does not exceed the annual digestive dose limit for the public at 0.290 mSv or 290 μSv

TABLE 6. Organ dose after the absorption of ^{238}U radionuclide in the human body and cancer risk assessment by type of cancer

Organ	Organ dose (μSv)	Life time cancer risk	Life time cancer risk (%)
Bones	65.95	2.31×10^{-4}	0.023
Liver	11.60	4.06×10^{-5}	0.004
Blood	8.41	2.94×10^{-5}	0.003

per year (UNSCEAR 2008). The risk assessment for fatal cancer based on the highest dose value in the study was also low at 1.109×10^{-5} or approaching 0.0011% alone 1 out of 100 000 people could get death from digestion of 2 g soil in the study area after 70 years. This value is lower than that reported by UNSCEAR (2000) that 1 out of 1000 people can get cancer. Based on the absorption of ^{238}U by the human body that occurs in the intestine, this study has identified three types of cancer that may be affected by the general public after 70 years of sampling of 2 g of soil samples. The highest probability of getting cancerous cancer is bone cancer, which is 0.023% followed by liver cancer by 0.004% and the latter is 0.003% of blood cancer. Overall, all the values for all the samples in this study include the specific concentration of ^{238}U activity, the annual effective dose and cancer risk assessment are at the safe level and below the prescribed standards.

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