

Use of RESRAD-Onsite 7.2 Code to Assess Environmental Risk around Tudor Shaft Mine Tailing Sites

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Abstract

The RESRAD-onsite 7.2 code has been used to assess the total dose rate in Tudor shaft site contaminated over an area of 10 km². The risk analysis simulation was span over a period of 1.0E+3 years. The maximum total dose of 1.64 mSv/yr was obtain at $t = 8.17 \pm 0.02$ years. The total peak dose at time $t = 0$ yr. is 1.63 ± 1.0 mSv/yr for all pathways. This value is 6.53 times higher compare to the basic radiation dose limit of 2.5E-01 mSv/yr. The evaluated excess cancer risk was 3.46E-3 and is 10 times higher compared to the recommended limit of WHO. A cover layer depth of 1.25 m was simulated using the code and a total maximum peak dose for all pathways was 2.52E-01 mSv/yr at $t = 5.0E+2 \pm 1.0$ years.

Keywords: RESRAD code, peak dose, distribution coefficient, excess cancer risk, contaminated site

1. Introduction

A primary consideration in formulating key environmental decisions and policies depends on potential risk to human health and the environment (Bellamy et al., 2014). Studying the possible impacts resulting from exposure to ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K often requires the modelling of the contaminant transport in the environment. According to literature the South African gold mines are associated with high levels of radionuclides. The mining activities in general tend to elevate the concentrations of NORMS near the earth surface (Njinga et al., 2016).

Tudor Shaft is an informal settlement in Krugersdorp which is west of Johannesburg and is highly contaminated by ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K due to the gold mine shaft and tailings dam. The community are affected by this mine's legacy. It is very obvious that people built their shacks on the tailings soil and grow their vegetables (resident farmer). There are children playing on the soils almost on daily bases which could result in soil ingestion. There are streams, wells and boreholes used by the population for irrigation, drinking, and household usages. The National Nuclear Regulator (NNR) in 2012, acting on advice from the local government began removing the Tudor Shaft waste dump. About half of the soil was removed, but environmentalists were alarmed that it was being done without risk-assessment studies or consultations.

²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K are common contaminants of concern due to its potential of high mobility in the environment. Dose assessment modelling of Tudor Shaft soil contamination with ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K would yield radiation dose results to indicate either a low soil clean up level that might require intensive remediation to attain, or a low waste acceptance criterion that would restrict the disposal of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K contaminated material in the soil (Gil-Garcia et al., 2009).

The RESRAD onsite code is developed by the U.S. Department of Energy (DOE) and the U.S. Nuclear Regulatory Commission (NRC) to evaluate radiologically contaminated sites (Yu et al., 2001a; Yu 1999, 2006, 2007). The code is used to derive clean-up criteria or Derived Concentration Guideline Levels (DCGLs) and estimate radiation dose or risk from residual radioactive material under various scenarios using appropriate parameters such as the cover materials with a total thickness, contamination depth, saturated zones, unsaturated zones, landfill spans an area and so on.

RESRAD have been widely used throughout the world for numerous benchmarking, verification, and validation studies. Recently, the U.S. Environmental Protection Agency (EPA) Office of Superfund Remediation and

Technology Innovation (OSRTI) issued a Memorandum (Office of Solid Waste and Emergency Response 9285.6-20) providing updated guidance on “Radiation Risk Assessment at Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) Sites (EPA 2014). The RESRAD code calculates both radiological dose and risk (NAS, 1999, Yu 1999, 2006, Walker, 2013). The dose assessments due ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K concentration in the soils would be analysed using RESRAD modelling for on-site exposures for the following pathways; external gamma, plant ingestion, meat ingestion, milk ingestion, drinking water and soil ingestion.

The aim of this study is to analyse the activity concentration of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K over an area of 10 km^2 and show how the k_{ds} values of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K for the loamy soil type contribute to the peak dose in the contaminated zone. It further evaluate the pathway contributions to the peak dose, when it occur over a span of 1000 years and calculate the excess lifetime cancer risk. Finally the study evaluates how the contaminated site could be clean using a clean cover layer of 1.25 m (depth) of soil so that the area would be use in farming and other basic activities and the people leave without any fear of health hazards.

2. Materials and Methods

2.1 The Study Area

The study area is in Krugersdorp ($26^{\circ}6'S$ and $27^{\circ}46'E$) and is a mining community in the West Rand District of Gauteng Province. The region covers a total area of 247.2 km^2 with about 140,643 people and population density of 570 persons per km^2 (Turton, 2004). In terms of geological arrangement, the West Rand area of Gauteng is underlain by rocks of the Johannesburg Subgroup of the Witwatersrand Super-group and is made up of erosion resistant quartzites, ironstones and some marine lava deposits interspersed with softer, more easily eroded tillites, mudstones and conglomerates and are high in water infiltration rate (Moore & Ramamoorthy, 1984). In some of the conglomerates of the younger members of the Super-group is found huge deposits of gold (Mendelsohn & Potgieter, 1986). Some of the minerals that have been associated with gold mining in the study area are pyrite and uranite. The map of the study area showing field sampling information is shown in Figure 1.



Figure 1. Map of the study area showing some field sampling information and the area of coverage

Gold mining has left more than 200 tailings dumps in the West Rand region where Krugersdorp is located for decades (Venter, 1995). The soil is composed mostly of sand with particle size $> 63\ \mu\text{m}$, silt with particle size $>$

2 μm , and a smaller amount of clay with particle size $< 2 \mu\text{m}$ and can be classified as loamy soil type. By weight, its mineral composition is about 40 % concentration of sand, 40 % concentration of silt and 20 % concentration of clay. The soil is suitable for growing most plant varieties.

2.2 Rate of Radionuclide Release from the Contaminated Zone

The release rate of radionuclides from the contaminated zone is estimated by using a nuclide dependent, first-order leach rate constant, which is defined as the fraction of available radionuclide i leached out per unit of time. The radionuclide release rate can be written as:

$$\frac{R_i(t)}{dt} = L_i \times \rho_b \times A \times T(t) \times S_i(t) \quad 1$$

where; L_i = leach rate for radionuclide i (yr^{-1}), ρ_b = bulk density of the contaminated zone (kg/m^3), A = area of the contaminated zone (m^2), $T(t)$ = thickness of the contaminated zone at time t (m), and $S_i(t)$ = average concentration of the i^{th} principal radionuclide in the contaminated zone available for leaching at time t .

The first-order leach rate constant used in RESRAD is time independent radionuclide leach rate constant. It is estimated on the basis of the soil residence time for the initial thickness of the contaminated zone and is written as;

$$L_i = \frac{I}{V_{wc} \times T_0 \times R_f} = \frac{(1 - C_e) \times [P_r \times (1 - C_r) + I_r]}{V_{wc} \times T_0 \times R_f} \quad 2$$

where; I = infiltration rate (m/yr), V_{wc} = volumetric water content of the contaminated zone defined as the product of the saturated water content of the contaminated zone V_{wc}^s and the saturation ratio of the contaminated zone R_s . The saturated water content V_{wc}^s is defined as the water content when the soil material is saturated. Hence, the saturated water content of the contaminated zone V_{wc}^s is equals to the total porosity P_t of the soil material.

T_0 = initial thickness of the contaminated zone (m), R_f = retardation factor in the contaminated zone for i -radionuclide, C_e = evapotranspiration coefficient, C_r = runoff coefficient (dependent on the environmental setting and the slope of the contaminated zone) P_r = precipitation rate (annual rainfall), and I_r = irrigation rate (m/yr).

When the medium is saturated, then the saturation ratio of the contaminated zone R_s equals unity. Under unsaturated infiltration conditions, the saturation ratio is a function of the infiltration rate, the saturated hydraulic conductivity, and the texture of the soil. The saturation ratio can be estimated by using the following equation (Clapp and Hornberger 1978):

$$R_s = \left[\frac{I}{H_C^s} \right]^{\frac{1}{2b+3}} \quad 3$$

where; H_C^s = saturated hydraulic conductivity (m/yr), b = soil-specific exponential parameter.

The retardation factor for radionuclide i , R_{f_i} is the ratio of the average pore water velocity to the radionuclide transport velocity. Assuming that the adsorption-desorption process can be represented with a linear isotherm; the retardation factor can be calculated with the following formula (Yu 1987, 1999):

$$R_{f_i} = 1 + \frac{\rho_b k_{d_i}}{V_{wc}} = 1 + \frac{\rho_b k_{d_i}}{P_t R_s} \quad 4$$

where; k_{d_i} is the distribution coefficient for the i^{th} principal radionuclide (cm^3/g).

From the above explanations, it is known that leach rate of a radionuclide is determined by its k_{d_i} value, which decides the relative transport speed of the radionuclide to that of water in the pore space. The leach rate also depend on the water infiltration rate, which determines the capacity of the liquid phase in soil, soil properties such as bulk density, porosity, saturated hydraulic conductivity, and the b-parameter (Eq 3) and the extent of contamination, which is described by contaminated zone thickness, area, and radionuclide concentration.

2.3 Hydrogeological and Hydro geochemical Properties

The hydrogeological and hydro geochemical properties of the zone that lie below the contaminated zone are assumed the same as those of the saturated stratum. The RESRAD code allows up to five horizontal strata below the contaminated zone, that is, $n \leq 5$. If $n = 0$, the contaminated zone extends down to the aquifer. The distance from the ground surface to the water table, $D_{wr}(t)$, at time t is evaluated using the relation below;

$$D_{wr}(t) = C_d(t) + T(t) + \sum_{m=1}^{n+1} \Delta z_m \quad 5$$

where; $C_d(t)$ = cover depth at time t (m), and $T(t)$ = thickness of contaminated zone at time t (m).

3. Materials and Methods

3.1 Collection of Soil Samples

The 145 soil samples were collected according to the internationally established experience as shown in Figure 1 over an area of 10 km². For each soil sample collected, an area of about 0.5 m × 0.5 m was marked and carefully cleared of debris to a few-meter depth (0.30 m) before collection of the soil samples. At the centre of each marked area, two kg of soil was collected using an auger at a depth of about 0.15 m from the ground so as to get the natural soil. Each soil sample was labeled according to the geographical coordinates of the sampling area. The 2 kg soil samples each making a total of 145 soil samples were transported to the Centre for Applied Radiation and Technology (CARST) laboratory at North-West University, South Africa, for analysis.

In CARST, the soil samples were crushed into fine powder using a mortar and pestle. The fine form of each soil sample was obtained using a scientific sieve of 0.2-mm mesh, dried in an oven at about 383 °K for 24 hours, hermetically sealed in standard 1500 mL plastic Marinelli beakers (assuming that radioactivity is homogeneously distributed in the whole volume of the measuring samples), dry-weighed and stored for about four weeks prior to counting to allow radioactive equilibrium among radon (²²²Rn), thoron (²²⁰Rn), and their short lived progenies. On average, 1.25 kg of soil was taken from each sample and put into 1.50 L Marinelli beakers for measurements using the HPGe detector.

3.2 Calibration of the Low Background Counting System

The calibration of the low background counting system was done using standard reference materials (SRM) from the International Atomic Energy Agency. The detector is a co-axial n-type and has a resolution of 2.0 keV at 1332 keV of ⁶⁰Co with a relative efficiency of 20 %. The output of the detector was analyzed using Canberra Genie 2000 software (Genie™ 2000). The detector is lead shielded to reduce the background level of the system (Xinwei and Xiaolon 2008). The efficiency calibration of the system was carried out using standard source of uranium ore in same geometry of our samples and the values plotted against energy for particular geometry. The full energy peak efficiency of a high purity Germanium (HPGe) detector is the number of gamma rays detected by the detector to the number of photons emitted by the source for a specific energy expressed mathematically as:

$$\varepsilon(E) = n(E) \times (A \times I_\gamma)^{-1} \quad 6$$

where, $n(E)$ = net count rate of the photo peak for the corresponding energy E , I_γ = Intensity of the gamma energy, A = present activities of the standard reference source.

For the evaluation of the efficiency of the detector (HPGe), the contributions for the coincidence summing effect, angular correlations due to the cascading gamma-rays were taken into account. The SRM uranium in silica matrix (RGU-1), thorium in silica matrix (RGTh-1) and potassium sulphate (RGK-1) with the same diameter as the soil samples of known concentrations of ²³⁸U, ²³²Th and ⁴⁰K radionuclides supplied by the Canada Centre for Mineral and Energy Technology (CAMET) under a contract with the IAEA were used for the calibration of detector efficiencies.

3.3 Activity Concentrations of Soil Samples

The radioactivity of each sample was measured using the calibrated HPGe-detector. Keeping the samples one by one on the top of the detector and counted for a period of 10,000 s the activity concentration of each radionuclide in the sample was determined by using the net count rates (Nc). After subtracting the background counts from the gross counts for the same counting time under the selected photo peaks, weight of the sample, the photo-peak

efficiency and the gamma intensity at a specific energy, a Computer software programming (GENE 2000) was used to analysed ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K .

The activity concentration of ^{226}Ra was determined using a photon peak of 609 keV (46.1%) from ^{214}Bi . Concentration of ^{232}Th was determined using the weighted mean of the gamma-ray transitions associated with the decays of ^{228}Ac , ^{212}Pb and ^{208}Tl . ^{238}U was determined from the average concentrations of the ^{214}Pb and ^{214}Bi decay products (Hamby and Tynybekov, 2000; Tzortzis et al., 2003). The ^{40}K concentration was determined using the gamma transition of 1461 keV (10.7%). Following the spectrum analysis, count rates for each detected photo peak and activity per unit mass (specific activity or radiological concentration) for each of the detected nuclides are calculated (Tzortzis et al., 2003). Thus, for each sample studied the specific activity concentrations of the ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K radionuclides are determined in units of Bq/kg according to the expression below (Olise et al., 2010):

$$A = \frac{C_{NP}}{BI \times \varepsilon(E_\gamma) \times m} \quad 7$$

where C_{NP} = net peak counts for a given energy line, BI = branching intensity, $\varepsilon(E_\gamma)$ = the absolute photo-peak efficiency of the detector and m is the mass of the sample in kg.

The calculation of external exposure from the specific activity concentrations of the ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K radionuclides determined in units of Bq/kg in RESRAD-Onsite 7.2 Code is based on the dose conversion factors and the formula obtained from the US Federal Guidance Report-12 (FGR-12) as published by Keith and Jeffrey (1993).

3.4 Quality Control

In order to perform quality control, a sample with known radionuclides concentrations were analysed using the same procedures. Based on the soil samples in this study the standard reference material IAEA-RGK-1, IAEA-RGTh-1, and IAEA-RGU-1 were analysed for using the gamma ray spectrometry system and the results compared in level of confidence with known radionuclides concentrations in the certificates.

3.5 Input Parameters

The radionuclide transformation was based on the International Commission on Radiological Protection-38 library (ICRP 2008), Federal Guidance Report (FGR) 11 & 12 for internal and external dose conversion library and FGR 13 library for health risk. The cut-off half-life was set to 180 days (6 months). This was based on the radionuclide in this study (^{238}U , ^{226}Ra , ^{232}Th and ^{40}K) with a cut-off half-life of six months as shown in Table 1.

Table 1. Principal and associated radionuclides with a cut-off half-life of 6 months

Principal Radionuclide ^a		Associated Decay Chain ^b
Species	Half-life (yr)	
^{40}K	1.28E+09	-
^{226}Ra	1.60E+03	^{226}Ra 1.60×10^3 ^{222}Rn (3.8235 d) ^{218}Po (3.05 min) [^{214}Pb (99.98%, 26.8 min) ^{218}At (0.02%, 2 s)] ^{214}Bi (19.9 min) [^{214}Po (99.98%, 1.64×10^{-4} s) ^{210}Tl (0.02%, 1.3 min)]
^{232}Th	1.41E+10	-
^{238}U	4.47E+09	^{234}Th (24.10 d) [$^{234\text{m}}\text{Pa}$ (99.80%, 1.17 min) ^{234}Pa (0.33%, 6.7 h)]

^a Radionuclides with half-lives greater than six months.

^b The chain of decay products of a principal radionuclide extending to (but not including) the next principal radionuclide or a stable nuclide. Half-lives are given in parentheses. Branches are indicated by square brackets with branching ratios in parentheses.

The use of site-specific parameter and defaults values obtained from literature were used for dose assessments (Yu et al., 2000) as shown in Table 2. However, the distribution coefficients (k_{ds}) of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K of the three zones (contaminated, saturated and unsaturated) were set to $15\text{ cm}^3/\text{g}$, $36000\text{ cm}^3/\text{g}$, $3300\text{ cm}^3/\text{g}$ and $55\text{ cm}^3/\text{g}$ respectively for the loamy soils to evaluate the transport mechanism to the water table.

Table 2. Basic defaults and site specific inputs values used in this study

Parameters	Site specific data	Default data
Area of contaminated zone	10 km ²	-
Thickness of contaminated zone	0.15 m	-
Cover depth	1.25 m	-
Length parallel to aquifer flow	100	-
Density of contaminated zone	1.5 g/cm ³	-
Contaminated erosion rate	-	0.001 m/yr.
Contaminated zone total porosity	-	0.4
Contaminated zone b- parameter	-	5.3
Evapotranspiration coefficient	-	0.5
Wind speed	2 m/s	-
Precipitation rate	1 m/yr	-
Irrigation rate	-	0.2 m/yr.
Density of saturated zone	1.5 g/cm ³	-
Saturated zone total porosity	-	0.4
Saturated zone effective porosity	-	0.2
Saturated hydraulic gradient	-	0.02
Saturated zone b- parameter	-	5.3
Water table drop rate	-	0.001 m/yr.
Well pump intake depth	10 m	-
Exposure duration	-	30 yrs.
Indoor time factor	0.5	-
Outdoor time factor	0.25	-
Fruits and grains consumption rate	-	160 kg/yr.
Leafy vegetable consumption rate	-	14 kg/yr.
Soil ingestion rate	-	36.5 g/yr.
Drinking water intake	-	510 liters/yr.

4. Results and Discussions

The analysis of ^{40}K , ^{226}Ra , ^{232}Th and ^{238}U in the standard reference material IAEA-RGK-1, IAEA-RGTh-1, and IAEA-RGU-1 with known activity concentration of respective reference values in Bq/kg evaluated by the gamma ray spectrometry system for quality control leads to good agreement for the radionuclides under validation. The percentage confidence were 95 % for ^{40}K , 97 % for ^{238}U , 98 % for ^{226}Ra and 86 % for ^{232}Th . As demonstrated by this technique the four radionuclides were analysed with good level of confidence. The results of the analysed soil samples for the Tudor shaft contaminated site are presented in Table 3.

Table 3. Results of activity concentration (Bq/g)

Radionuclides	Mean values	Range	Median
^{40}K	0.035 ± 0.002	0.023 - 0.039	0.025
^{226}Ra	0.915 ± 0.016	0.690 - 1.287	0.854
^{232}Th	0.018 ± 0.001	0.009 - 0.021	0.013
^{238}U	1.404 ± 0.061	0.974 - 1.898	1.215

The total peak dose at time $t = 1$ yr. due to ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K were $3.35\text{E-}02$ mSv/yr, 1.59 mSv/yr, $3.16\text{E-}03$ mSv/yr and $5.86\text{E-}03$ mSv/yr. The maximum total dose (t-max-D) of 1.64 mSv/yr was obtain at $t = 8.17 \pm 0.02$ yrs. The meat, milk and soil pathways total dose contributions were $1.496\text{E-}02 \pm 0.0091$, $1.547\text{E-}02 \pm 0.0094$, and $1.838\text{E-}02 \pm 0.0112$ mSv/yr respectively. The DCGLs values at $t = 8.17 \pm 0.02$ yrs were calculated

based the basic radiation dose limit (BRDL) of 0.25mSv/yr multiplied by the initial contaminant levels (Bq/g) of the radionuclides divided by the respective maximum total peak dose at time $t = 8.17 \pm 0.02$ yrs. The total DCGLs values at time of maximum total dose (t-max-D) were 3.423 Bq/g for ^{238}U , 1.421E-01 Bq/g for ^{226}Ra , 1.647E-01 Bq/g for ^{232}Th and 1.431 Bq/g for ^{40}K .

As seen in Figure 2, the total peak dose at time $t = 1$ yr. is 1.63 ± 1.0 mSv/yr for all pathways summed and is 6.53 times higher compare to the basic radiation dose limit of 2.5E-01 mSv/yr.

DOSE: All Nuclides Summed, All Pathways Summed

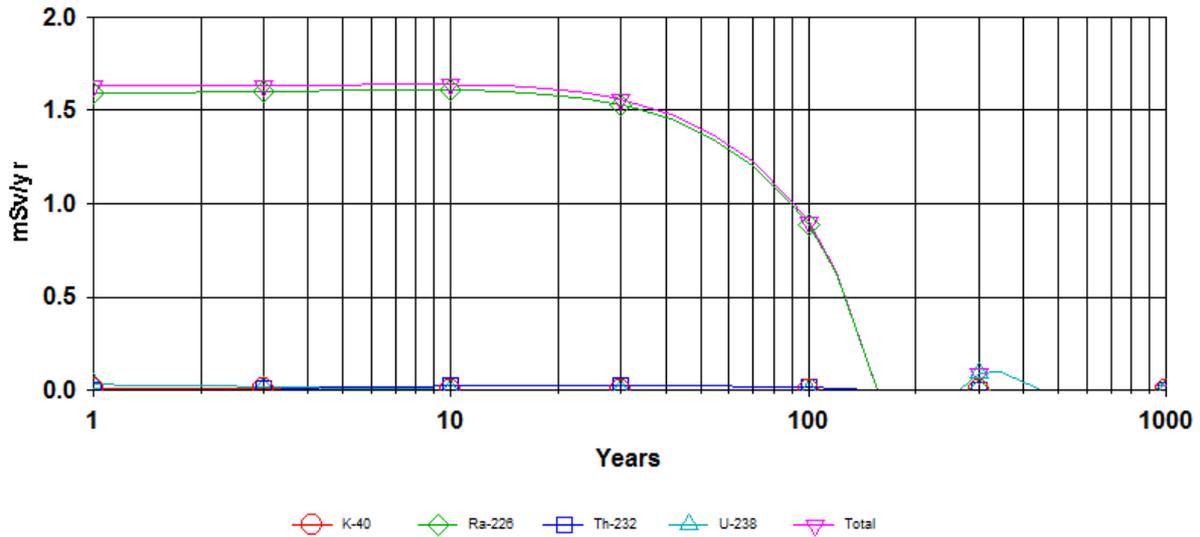


Figure 2. Summed total dose due to all nuclides and all pathways

The evaluated excess cancer risk as revealed in Figure 3, was observe to be constant from the 1st to the 10th year with approximate value of 3.46E-3. The value is 10 times higher compared to the recommended limit of WHO.

EXCESS CANCER RISK: All Nuclides Summed, All Pathways Summed

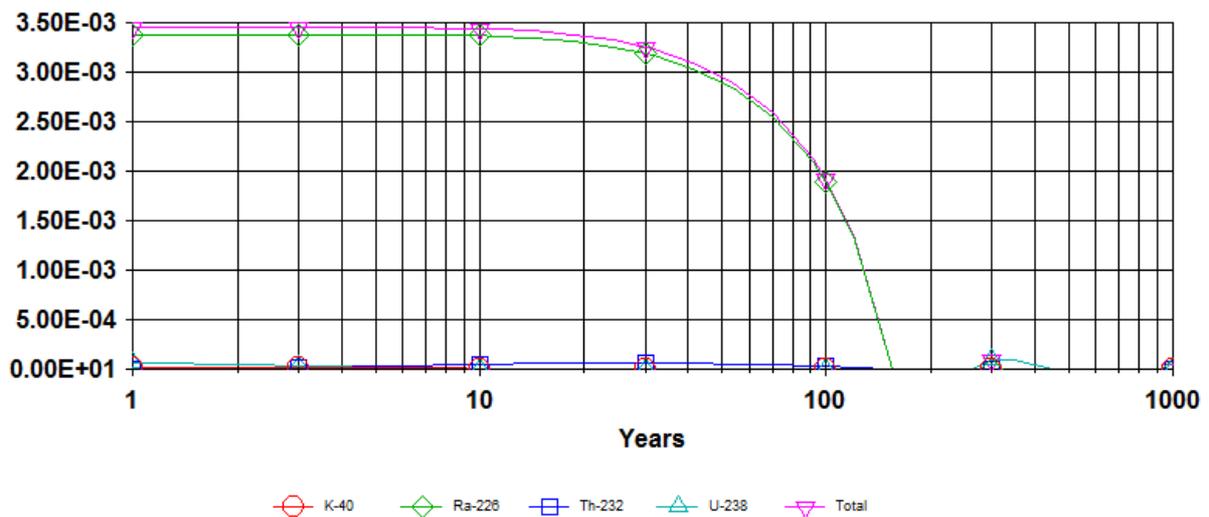


Figure 3. A log-scale plot of excess cancer risk, time, and nuclide contributions

As seen in Figure 2, ^{226}Ra is the major contributor (99.1%) at the early times to the excess cancer risk with a total value of 3.38×10^{-3} , which is 10 times the limit of 3.00×10^{-4} , obtained using recommended value of 0.25

mSv/yr. At about 11 years and above, there was a slow drop in the risk values up to the 100 yrs after which the risk was totally below the BRDL estimate.

In the RESRAD modelling, a transfer function is used which calculates the amount of progeny radionuclide in groundwater as observed at a certain time. This may be caused by the existence of parent radionuclide in the contaminated zone at time 0 to 10 yrs since ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K are transported in the soils. During these periods, the decay products (progeny radionuclides) also have the potential to dissolve and transported to deeper soils.

The early pathway as observed in Figure 4 is external gamma with a value of 1.39 mSv/yr at time t = 1 yr and later followed by plant ingestion (water independent) with a value of 0.42 mSv/yr. Later in the future, there was a drop in contribution from all the pathways considered in this study.

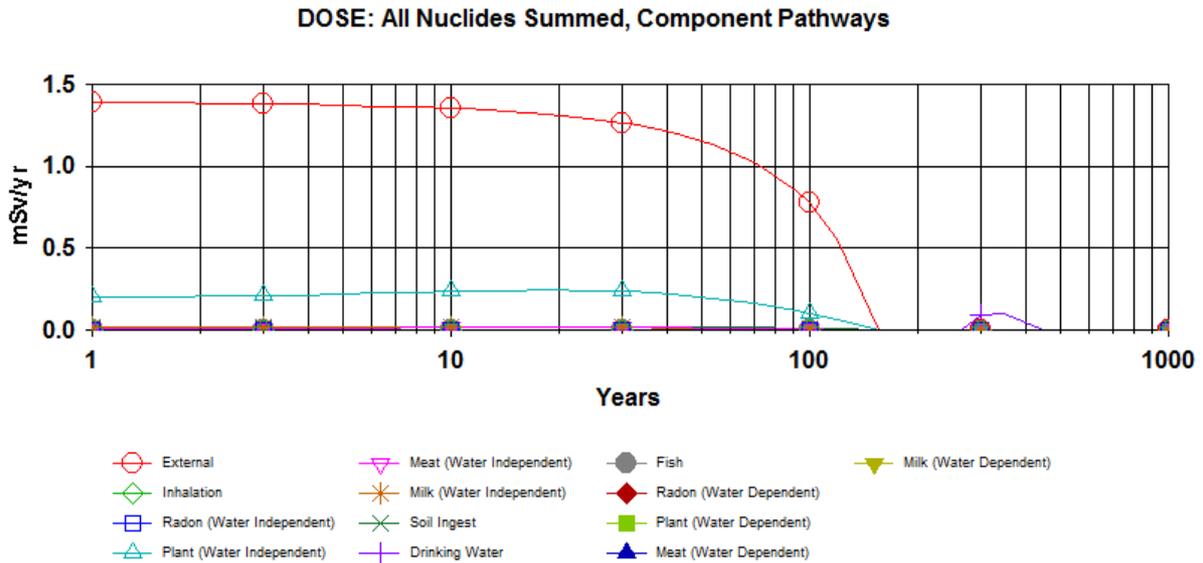


Figure 4. Log-scale plot of the pathway contributions to peak dose

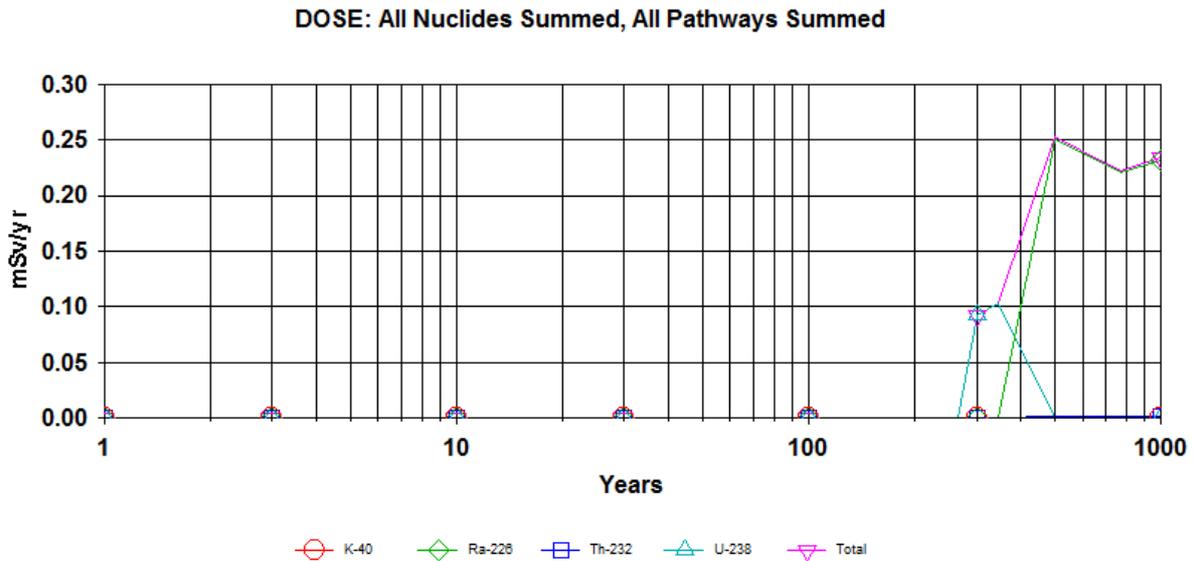


Figure 5. The summed dose due to all nuclides after a clean cover thickness of 1.25 m was used

The clean cover thickness of 1.25 m was used to calculate the level of clean-up of the contaminated site (Tudor Shaft). Based on this clean up cover thickness, the maximum total dose of 2.52E-01 mSv/yr was observed at t = 500 ± 1 years. As seen in Figure 5, the contaminated site under investigation with initial total maximum dose of

1.63 ± 1.0 mSv/yr for all pathways has been cleaned to a dose lower to 0.001 mSv/yr for a period of 1 to 500 yrs before it peak at 500 yrs to a dose ($2.52E-01$ mSv/yr) compared to the basic radiation dose limit (BRDL) of 0.25mSv/yr.

5. Conclusion

The RESRAD-onsite 7.2 code has been used to complete environmental risk assessment in Tudor shaft contaminated site of an area of 10 km^2 . The maximum total dose of 1.639 mSv/yr was obtain at $t = 8.17 \pm 0.02$ years with the meat, milk and soil pathways-total dose contributions of $1.496E-02 \pm 0.0091$, $1.547E-02 \pm 0.0094$, $1.838E-02 \pm 0.0112$ mSv/yr respectively. The total peak dose at time $t = 0$ yr. is 1.63 ± 1.0 mSv/yr for all pathways and is 6.53 times higher compare to the basic radiation dose limit of $2.5E-01$ mSv/yr with ^{226}Ra contributing 99.1%. The total excess cancer risk value of $3.46E-3$ is 10 times the limit of 3×10^{-4} obtained using BRDL. A cover clean-up layer of 1.25 m was used to achieve lower dose of 0.001 mSv/yr for a period of 1 to 500 yrs before it peak at 500 yrs to a dose ($2.52E-01$ mSv/yr) compared to the basic radiation dose limit (BRDL) of 0.25mSv/yr.

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