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Radiological Atmospheric Risk Modelling of NORM Repositories in Hungary

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Abstract: The human population is continuously exposed to natural radionuclides in environmental elements. The concentration of these nuclides is usually low, but different technological processes and activities can concentrate them in products, by-products, or wastes. These activities are, for example, coal mining, fertilizer production, ore mining, metal production, etc. These materials are labelled as NORM (Naturally Occurring Radioactive Material). The most common method of disposal for NORMs is deposition in different types of depositories. The long-term effects of these depositories on the environment and on human health are hard to estimate. The aim of the study is to assess radiation risk from the five selected NORM depositories (Ajka coal ash, Ajka red mud, Almásfüzitő red mud, Zalatárnok drilling mud, and Úrkút manganese residue) for members of the public and biota. The radionuclide concentrations were determined by HPGe gamma-spectrometry. The measured concentration was between 31 Bq/kg and 1997 Bq/kg for Ra-226, between 33 Bq/kg and 283 Bq/kg for Th-232, and between 48 Bq/kg and 607 Bq/kg for K-40. The dose estimation was investigated using RESRAD-ONSITE and RESRAD BIOTA, which are computer codes developed by the Argonne National Laboratory (USA). RESRAD-ONSITE can estimate the radiation risk from the radionuclides in the contaminated sites. The highest dose was observed in the case of the Ajka coal ash depository–without cover (12.38 mSv/y), and the lowest was in the case of Zalatárnok (0.53 mSv/y). The most significant contributors to the population dose are the uptakes through plants and external pathways, which account for more than 80% of the total dose on average. RESRAD-BIOTA code was used to estimate the radiation exposure of terrestrial organisms (plants and animals). During this work, the values of sum ratio factor (SRF), biota concentration guide (BCG), external dose, internal dose, and total dose were determined.

Keywords: NORM repositories; RESRAD-ONSITE; RESRAD-BIOTA; atmospheric modelling; radiological assessment

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1. Introduction

The population is constantly exposed to ionising radiation from natural and artificial sources [1]. The background radiation of natural origin is mainly from primordial radionuclides and their decay products. The concentrations of these isotopes in some environmental elements are not very high in general, but in certain areas and in some mineral formations, significant levels can be observed [2–5]. Those materials with much higher-than-average concentrations of natural isotopes are called NORM (Naturally Occurring Radioactive Material) [6,7]. These higher concentrations can occur in natural conditions (e.g., caves), but also as a result of human activities such as mining [8,9], and bauxite processing [10,11].

The management, disposal, and possible use of bottom ash from coal use have long been a serious problem [12]. Several studies have been carried out to investigate its radiological aspects [13,14]. The radionuclide content of coal varies over a wide range [15,16], and consequently, the amounts of these isotopes present in the bottom ash will also vary [17]. In the past, the by-products were used in the construction industry without restrictions, but this caused problems in many cases (e.g., Tatabánya).

Red mud is the processing of bauxite into aluminium dioxide, an industrial waste from the Bayer process. It is highly chemically contaminated, and its radionuclide content depends mainly on the radionuclide content of the bauxite [18]. Its recyclability is severely limited and it is currently disposed of in permanent storage [19,20]. Dewatering following the drying of the slurry, which in many cases can contaminate surrounding areas with radionuclides, can be a significant problem.

Several by-products are generated during the extraction and processing of oil [21]. In some of these, radionuclides extracted with the oil can accumulate and even develop elevated concentrations. The extent of the accumulation depends on the stage of the processing.

Mining activities generate a large quantity of waste rock, most of which is disposed of in surface storage. These tailings contain radionuclides at different levels depending on the type of ore mined. This is because the processes that take place during the formation of the ore favor the accumulation of certain elements. This can happen, for example, when manganese ores are formed [22,23].

The currently accepted management of these industrial by-products of human activities is to place them in repositories. However, this has the drawback that the accumulated isotopes can cause significant long-term exposure, to both external and internal loads.

The external radiation exposure is mainly short-term and can be reduced by covering and recultivating the repositories.

Internal radiation exposure is less significant in the short term unless there is a significant amount of dust from the landfill. In the long term, however, radioisotopes can be transported via plants into the food chain through mobilisation, ultimately into the human body.

RESRAD is a series of computer codes developed by Argonne National Laboratory (USA) for environmental risk assessment. It can analyse the human and non-human radiation exposures and estimate the radiation risk from the contamination of **residual radioactive** materials. RESRAD has five main subtypes: RESRAD-ONSITE, RESRAD-OFFSITE, RESRAD-BUILD, RESRAD-RDD and RESRAD-BIOTA. The program can be applied to the performance of a pathway analysis to estimate the exposure and radiation risk or to determine the cleanup criteria for radionuclides in contaminated areas [24]. The RESRAD codes are widely used by regulatory agencies, research institutes, and universities in more than a hundred countries around the world.

The aim of this study is to estimate the radiological risk from some NORM repositories that had been investigated previously [11,25–27]. For this purpose, the activity concentrations of Ra-226, Th-232, and K-40 in NORM materials were taken as a basis. An estimation of the external exposure from uncovered repositories was performed by running the RESRAD-ONSITE code, while the exposure of the non-human biota was investigated by using the RESRAD BIOTA program.

2. Materials and Methods

2.1. Study Areas

Five NORM depositories were selected for radiological atmospheric risk modelling. The depositories' locations and distribution are indicated in Figure 1.

Ajka coal ash depository site: As a result of the high radioactivity of deposited ashes in this site, due to the high Ra-226 content in coal around Ajka, this location was considered for this study.

Red mud depository sites: Due to the ecological and radiological hazard of red mud to the environment, two sites, namely "Ajka" and "Almásfűzitő", were selected for further investigation.

Zalatárnok residue depository site: Residues and mud from oil extraction in Zala County (Hungary) are deposited near Zalatárnok, which makes it important for radiological investigation due to the residues' properties as a result of concentrated radionuclides accumulated in the residues.

Úrkút manganese mud depository site: This site was considered as the depository materials in this site typically show potential for U-rich formations.

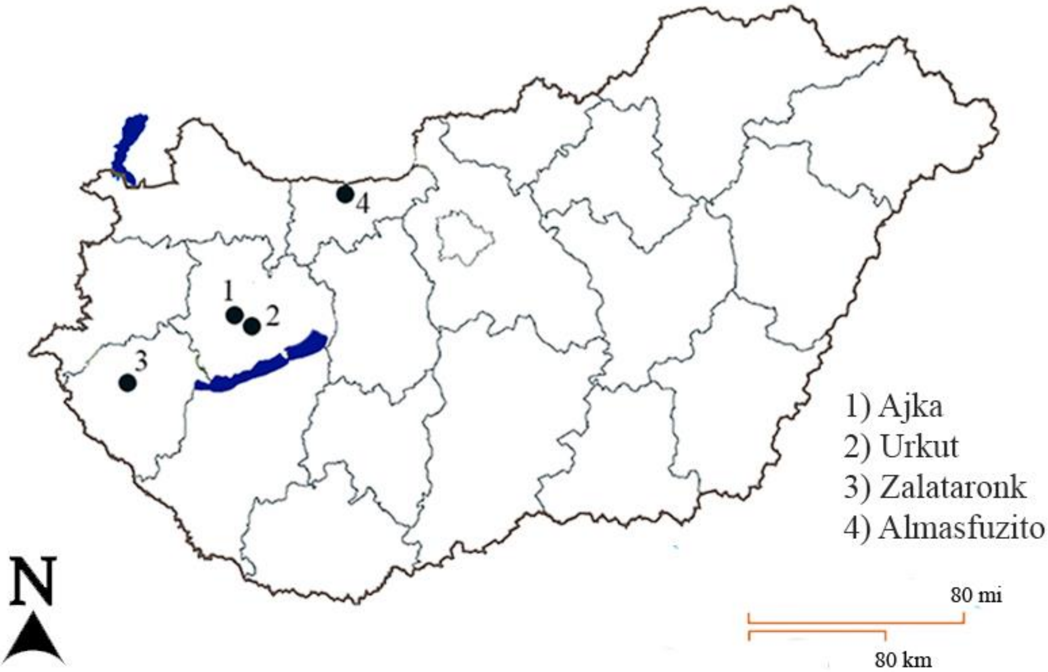


Figure 1. Map of Hungary presents the five investigated depositories.

2.2. Determination of Terrestrial Radionuclides' Concentration

As this paper mainly deals with radiological risk modelling and analysis, discussion about gamma dosimetry and radiological measurements is out of its scope. Therefore, the methodology for activity concentration of radionuclides measurements, uncertainty, and minimum detectable activity (MDA) was determined using calculations explained in previously published papers. More information regarding gamma spectrometry can be found in the previously published papers [7,11,28].

2.3. RESRAD-ONSITE

RESRAD-ONSITE (Version 7.2.) was applied to estimate the cancer risk from the natural radionuclides in NORM storage.

Several radiological parameters (including the contamination's characteristics, surface, sub-surface, and saturated soil parameters, site-specific meteorological, hydrological, and hydrogeological properties, and the exposure pattern of the receptor) were considered for this study, which are shown in Table 1. The RESRAD-ONSITE modelling accounts for radiological decay and in-growth, as well as environmental transport, partitioning, and dilution, controlled by the principle of conservation of mass over time. Basically, all input variables used for the calculation can be defined based on a study by the user; therefore, the operator can specify the level of conservatism of each analysis and apply the RESRAD-ONSITE code for screening, site-specific screening, or site-specific evaluation tasks.

The estimation of dose and cancer risk by the RESRAD-ONSITE code is scenario-based, using user-defined parameter values. There are nine exposure pathways which can be selected or blocked according to the land use and receptor scenario under examination.

These pathways, considering the outdoor condition, are: (1) direct external radiation, (2) inhalation of radionuclides in the air that are resuspended or evaporated, (3) ingestion of soil, (4) ingestion of plant foods grown in contaminated areas and watered with contaminated water, (5) ingestion of meat, (6) consumption of milk produced by animals fed with contaminated feed and water, (7) drinking water from a well or pond in the vicinity of the contaminated site, and (8) ingestion of aquatic foods. The modelling requires input parameters including the contamination's characteristics, surface, sub-surface, and saturated soil parameters, site-specific meteorological, hydrological, and hydrogeological properties, and the exposure pattern of the receptor. The RESRAD-ONSITE modelling accounts for radiological decay and in-growth, as well as environmental transport, partitioning, and dilution, controlled by the principle of conservation of mass over time. Basically, all input variables used for the calculation can be defined by the user; therefore, the operator can specify the level of conservatism of each analysis and apply the RESRAD-ONSITE code for screening, site-specific screening, or site-specific evaluation tasks [29].

The selected reservoirs for this study are all covered, except for the red mud reservoirs. The modelling was carried out without cover and with 1 m cover.

Table 1. Site-specific parameters.

	Coal ash	Red Mud (Ajka)	Red Mud (Almásfüzitő)	Drilling Mud	Manganese Residue
Area of contaminated zone [m^2]	10,000	300,000	1,720,000	17,000	18,100
Thickness of the contaminated zone [m]	10	11	10	2	6
Cover depth	0	0	0	0	0
Density of the contaminated zone [g/cm^3]	2.4	2.7	2.7	1.0	4.5
Wind speed [m/s]	4.25	4.25	2.75	2.25	4.75
Precipitation rate	0.725	0.725	0.575	0.775	0.725
Indoor time factor	0.6	0.6	0.6	0.6	0.6
Outdoor time factor	0.4	0.4	0.4	0.4	0.4

2.4. RESRAD-BIOTA

RESRAD-BIOTA code is for estimating radiation dose to nonhuman, aquatic, and terrestrial biota [30]. Radiation exposure to living organisms in terrestrial or aquatic ecosystems comes from contaminated soil, water, and sediment, which then pollute the air and various food sources. The RESRAD-BIOTA code uses a stepwise approach with three levels of analysis. The Level 1 screening BCGs (Biota Concentration Guide) for contaminated soil, water, and sediment, developed by DOE (Department of Energy, Washington, DC, USA) using four default organism categories, terrestrial animals, terrestrial plants, riparian animals, and aquatic animals, are used to compare input concentrations of environmental media to determine whether the recommended biotic dose limit may be exceeded. At Levels 2 and 3, more site- and organism-specific input data are considered to make a more accurate dose calculation for comparing with the determined dose limit. Both external and internal radiation are taken into account in the dose calculation. The calculation of the external dose should take into account the periods of time that the organism spends near or in the contaminated medium. Three options are available for calculating the internal dose: measured tissue concentrations, the medium to tissue concentration ratio, or allometric equations that assess the maximum tissue concentration, taking into account inhalation and uptake rates, biological and radiological decay, body mass and organism lifetime [29].

In this study version 1.8. was applied to determine the BCG value, total dose, and sum ratio factor. Among the terrestrial plants, grass was studied, while among the animals, cows and deer were selected. The average weight was chosen to be 800 kg (normal Hungarian cows weigh from 650 to 900 kg) for cows, and 150 kg for deer (normal Hungarian deers weigh from 90 to 200 kg).

Biota Concentration Guide (BCG) is the critical concentration of radionuclides in the soil at which the dose level criteria for the protection of terrestrial biota would not be exceeded. The determination for the radionuclide i is given by Equation (1).

$$BCG_{\text{soil, terrestrial plant/animal, i}} = \frac{365.25 \cdot DL_{tp/ta}}{CF_{tp/ta} \cdot [(B_{iv,tp/ta,i} \cdot DCF_{int,i}) + DCF_{ext,i,sol}]} \quad (1)$$

where BCG soil, terrestrial plant/animal, i is the radionuclide i concentration in the soil [Bq/kg]; $DL_{tp/ta}$ is the recommended dose limit for terrestrial plant/animal [0.01 Gy/d for plant and 0.001 Gy/d for animal); $CF_{tp/ta}$ is the correction factor for area or time (default value is 1); $B_{iv,tp/ta,i}$ is the fresh mass terrestrial plant/animal to soil concentration factor of radionuclide i; $DCF_{int,i}$ is the dose conversion factor to assess the dose rate of the issues form radionuclide i $[\frac{Gy/y}{Bq/kg}]$; $DCF_{ext,i,soil}$ is the dose conversion factor to assess the external dose rate.

The Sum Ratio Factor (SRF) is the absorbed dose in the biota relative to the total dose limit. The dose rate limit is 0.01 Gy/d for terrestrial plants and 0.001 Gy/d for terrestrial animals [31,32].

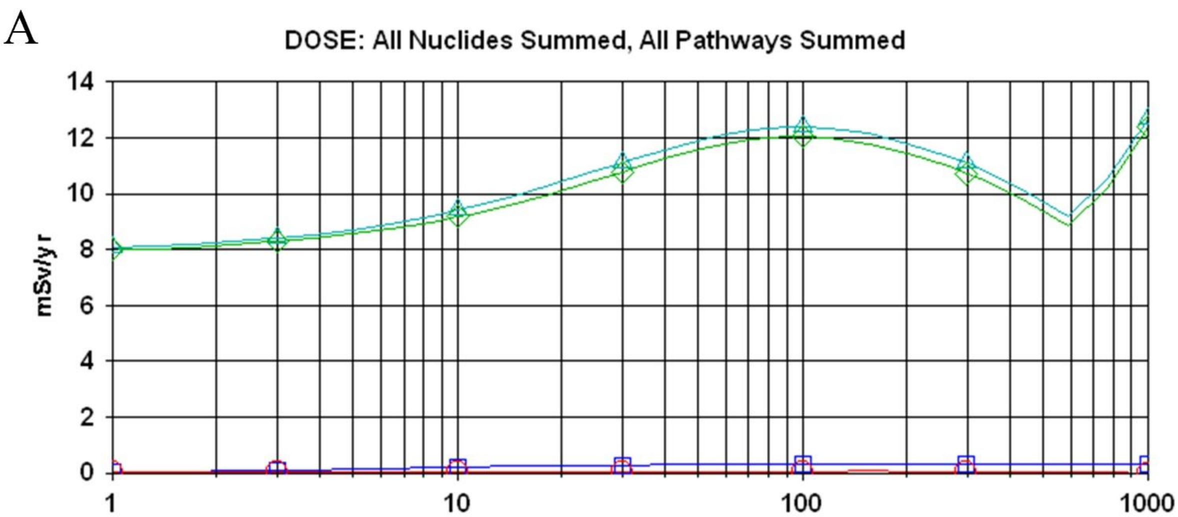
3. Results

The results from gamma-spectrometry measurement are shown in Table 2. The measured concentration was between 31 Bq/kg and 1997 Bq/kg for Ra-226, between 33 Bq/kg and 283 Bq/kg for Th-232, and between 48 Bq/kg and 607 Bq/kg for K-40. The minimum detectable activity of the same radionuclides, based on the observed data from background measurement, was calculated at 23, 0.5, and 0.7 Bq/kg, respectively [28].

Table 2. Activity concentration of the radionuclides in the investigated NORMs.

Investigated NORM	Ra-226 [Bq/kg]	Th-232 [Bq/kg]	K-40 [Bq/kg]
Coal ash (Ajka)	1997	33	56
Red mud (Ajka)	182	245	284
Red mud (Almásfüzitő)	347	283	48
Drilling mud (Zalatárnok)	31	35	502
Manganese residue (Úrkút)	52	40	607

Risk and dose estimates for NORM reservoirs were determined using the RESRAD-ONSITE code. Figure 2 shows the magnitude of annual doses from uncovered storage over a 1000-year period. The maximum annual dose value, the observation of the year, and the contribution of Ra-226, Th-232, and K-40 to this dose are shown in Table 3. The highest value was calculated for the Ajka coal ash deposit (12.38 mSv/year).



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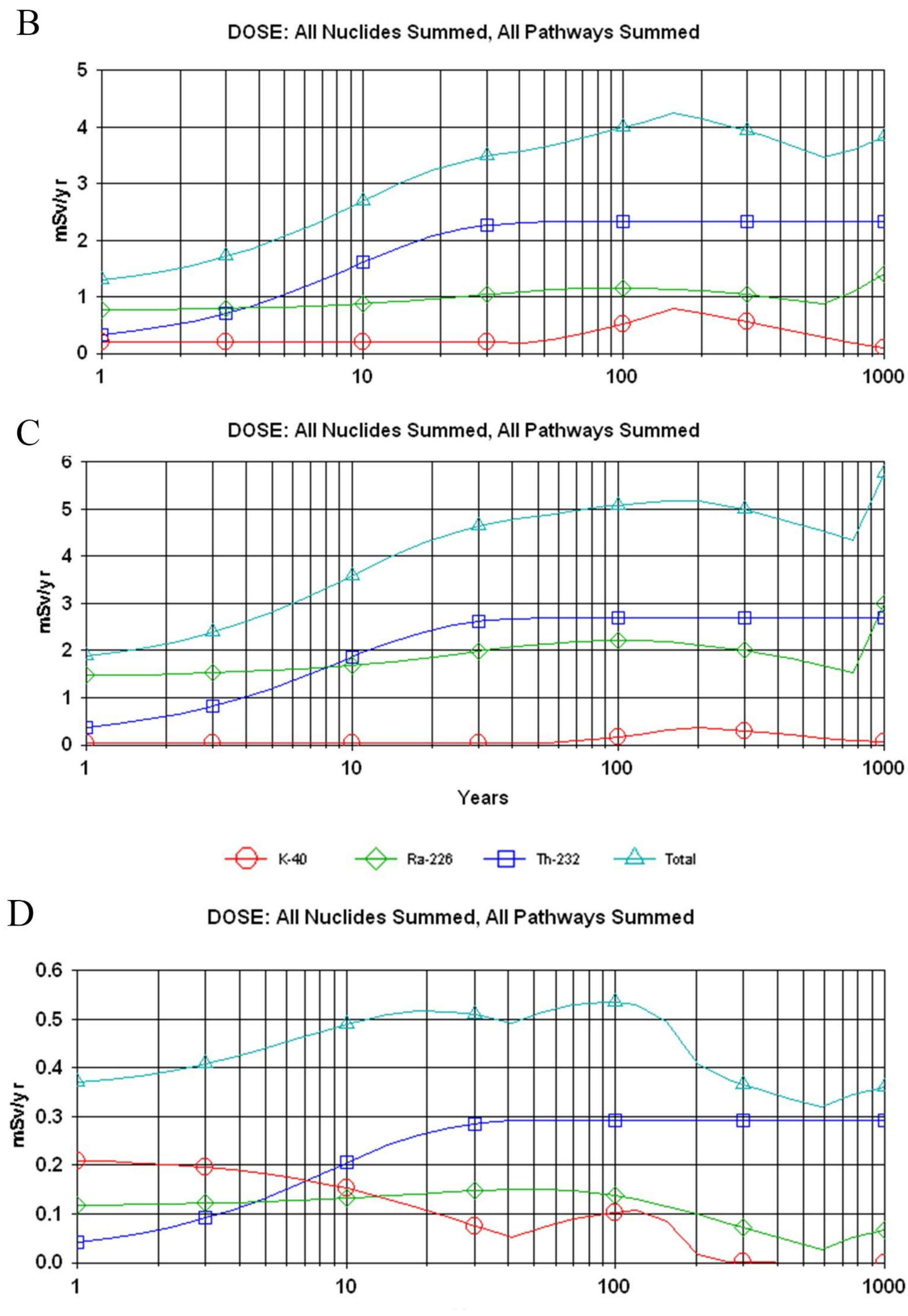

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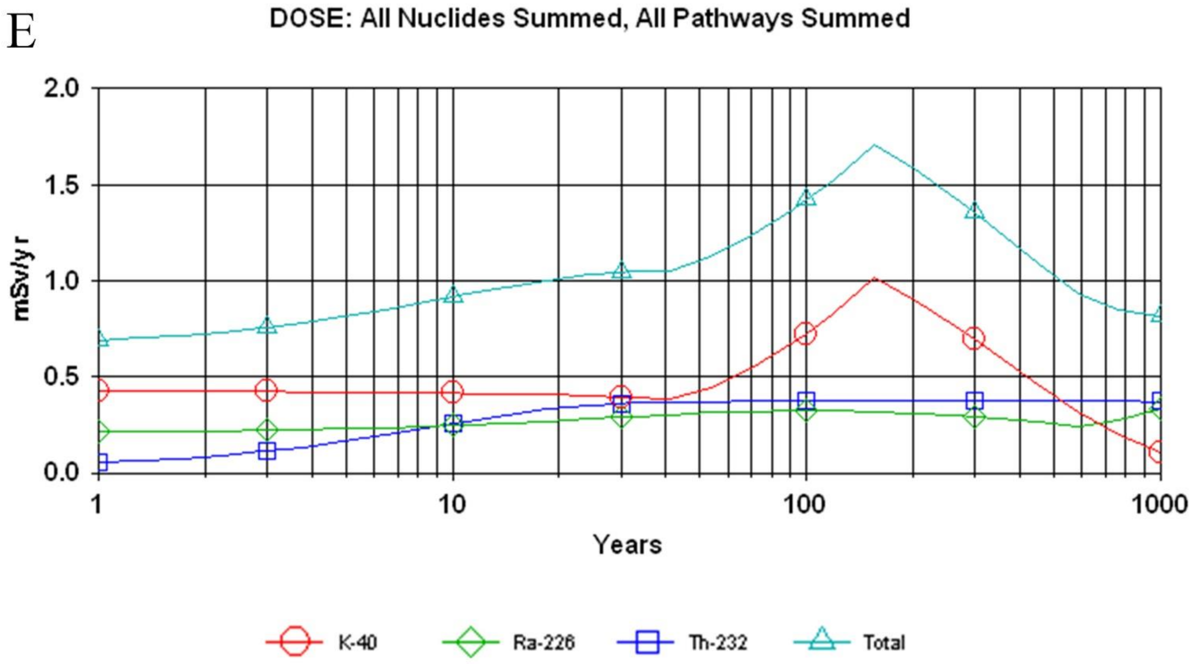


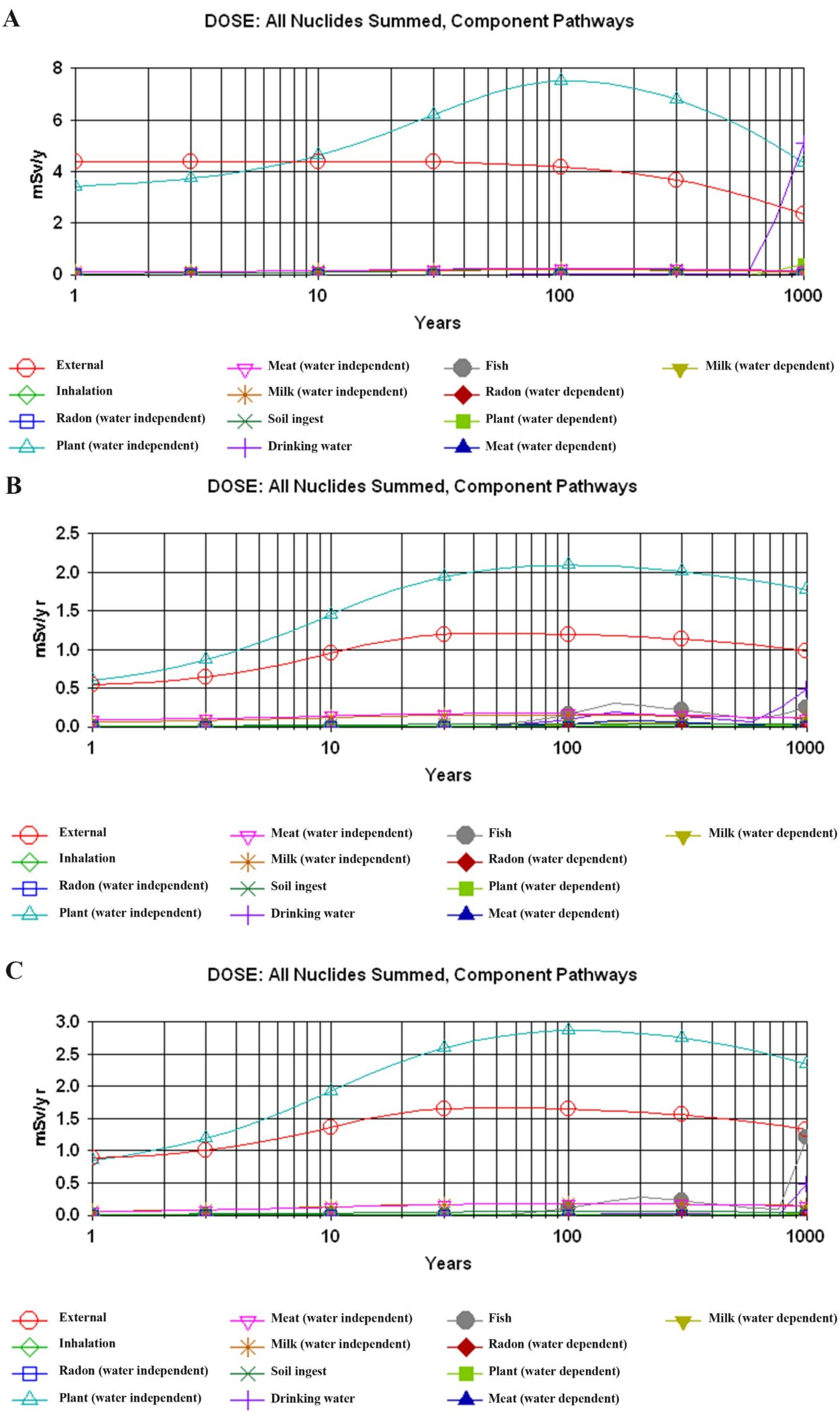
Figure 2. Total annual dose from all radionuclides and all pathways. (**A**) Ajka coal, (**B**) Ajka red mud, (**C**) Almásfüzitő red mud, (**D**) Zalatárnok drilling mud, (**E**) Úrkút manganese.

Table 3. The maximum annual dose value, the observation of the year, and the contribution of Ra-226, Th-232, and K-40 to this dose.

Investigated NORM Depository	Maximum Annual Dose [mSv/y] (Year of Observation)	Contribution of the Different Radionuclides [mSv/y]		
		Ra-226	**Th-232**	**K-40**
Ajka–coal ash	12.38 (t = 100 year)	12.03	0.30	0.05
Ajka–red mud	4.25 (t = 156 year)	1.13	2.32	0.80
Almásfüzitő–red mud	5.18 (t = 156 year)	2.18	2.70	0.30
Zalatárnok-drilling mud	0.53 (t = 100 year)	0.14	0.29	0.10
Úrkút-manganese residue	1.71 (t = 156 year)	0.32	0.37	1.02

The distribution between the exposure pathways is shown in Figure 3. The most significant contribution comes from uptake through plants, followed by the external pathway. These two account for more than 80% of the total dose on average. The only difference from this is seen at the Ajka coal slag reservoir, where the external pathway was initially the most significant (more than half of the total dose), then reverses at t = 8 years and around t = 1000 years of drinking water becomes the most important contributor.

Figure 4 shows cancer risk estimated by the REDRAD-ONSITE code. The shape of the curve is similar for all storage. Initially, there is a growth phase with a maximum between 100 and 200 years (in the range of 4.15×10^{-4} to 1.81×10^{-2} for total risk). This is followed by a fall and then a rise again at the end (t = 1000 years). Since the modelling considers the radiation exposure and risk from decay products, the increase is due to the mobilization and availability of these progenies. The magnitude of change varies from one landfill to another. The highest risks are Ra-226 for coal slag (1.76×10^{-2}), Th-232 for red mud (3.45×10^{-3}), and K-40 for manganese (2.87×10^{-3}). For drilling mud reservoirs, thorium and potassium are the dominant nuclides, with the nuclide posing the greater risk varying with time.



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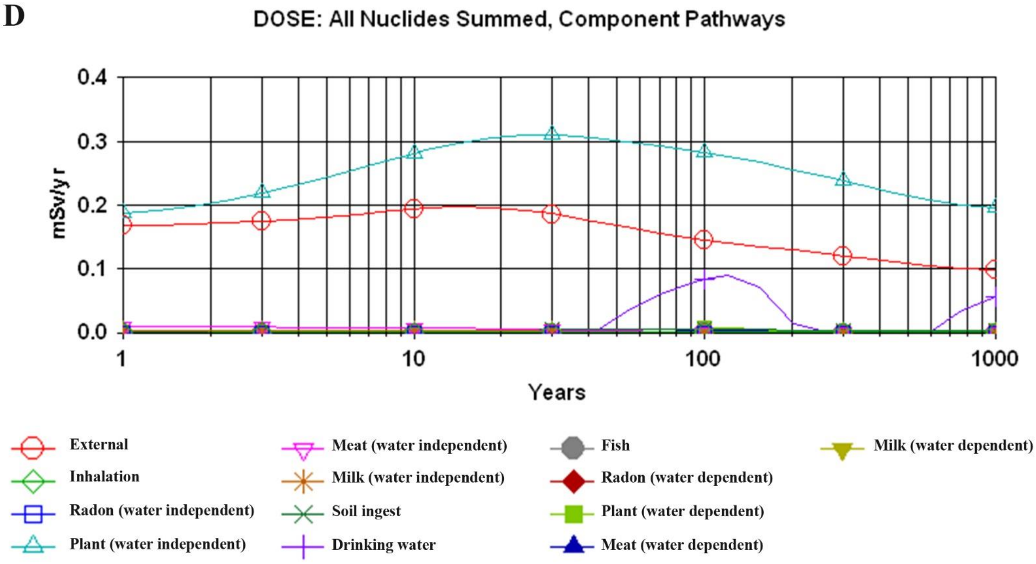

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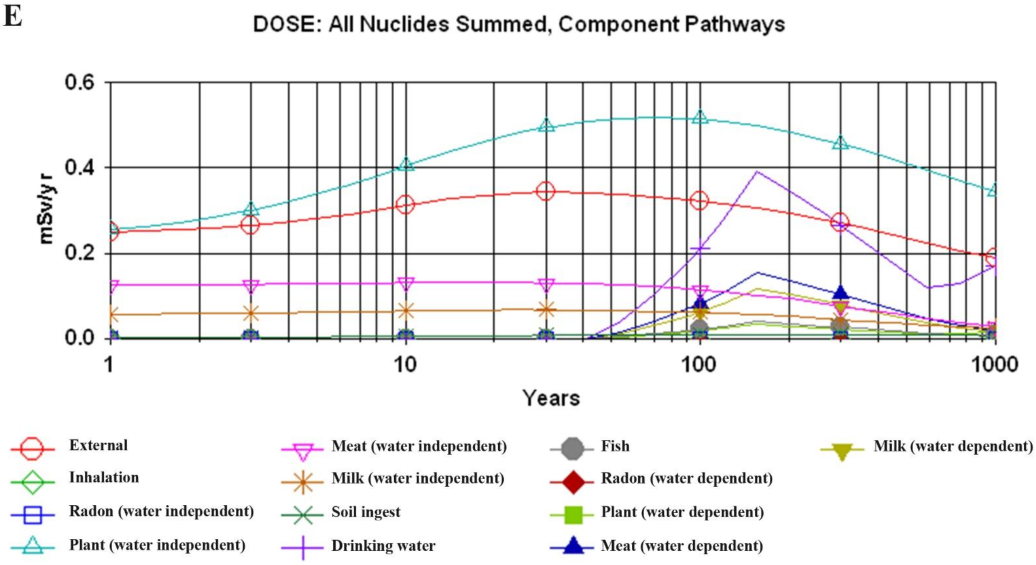
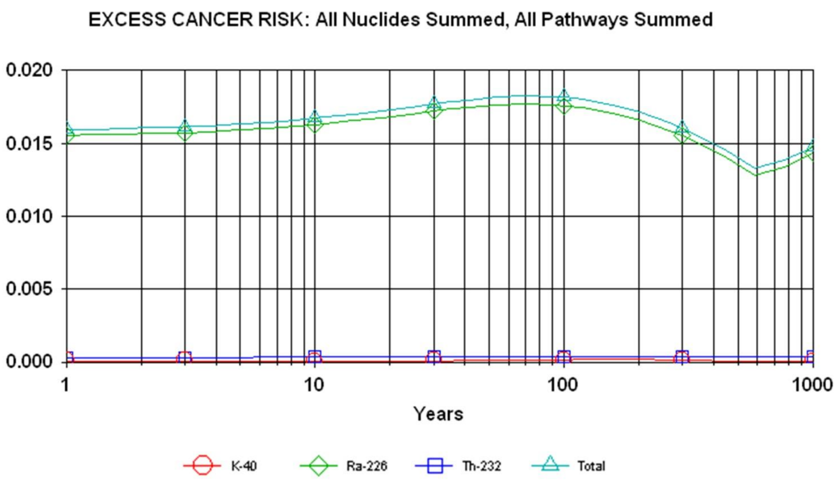


Figure 3. Contribution of different exposure pathways to total dose. (**A**) Ajka coal, (**B**) Ajka red mud, (**C**) Almásfüzitő red mud, (**D**) Zalatárnok drilling mud, (**E**) Úrkút manganese.

A



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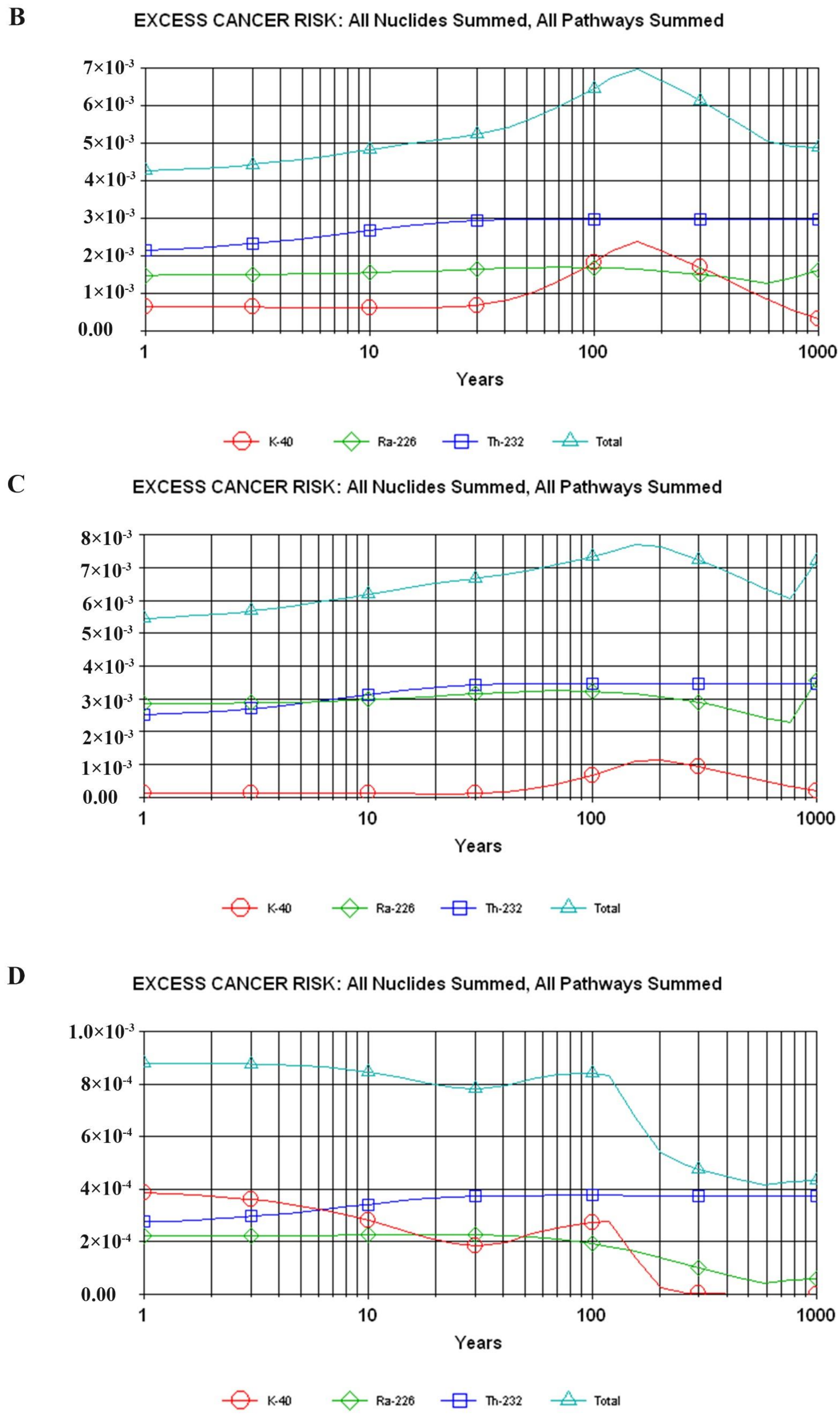

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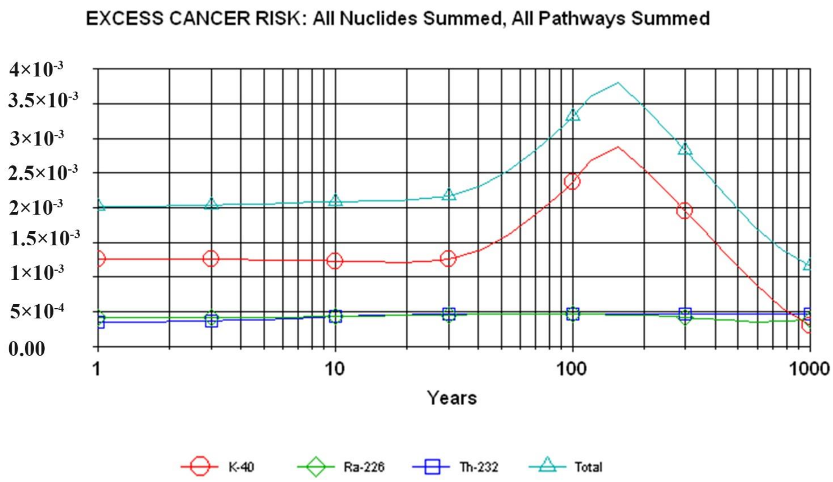
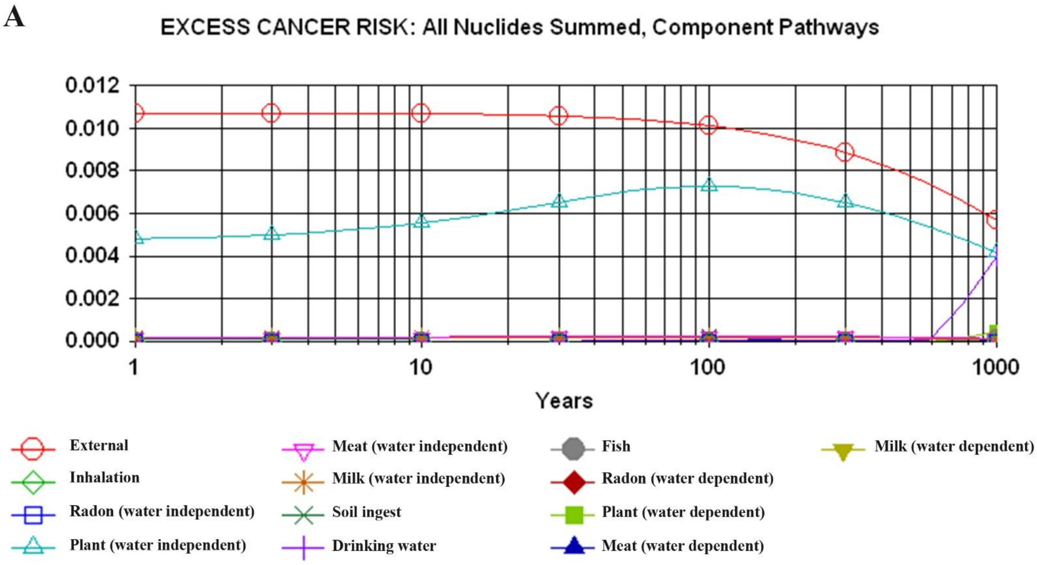


Figure 4. Cancer risk from all radionuclides. (**A**) Ajka coal, (**B**) Ajka red mud, (**C**) Almásfüzitő red mud, (**D**) Zalatárnok drilling mud, (**E**) Úrkút manganese.

Figure 5 shows the distribution of cancer risk between different exposure pathways. Although in the case of dose, the contribution of plants was the dominant contributor, closely followed by external exposure, in case of the cancer risk external exposure is the most significant contributor. The risk from external exposure is nearly constant or increases slightly for the first 100 years, then starts to decrease after 100 years. The rate of reduction is different for each storage.

Figure 6 shows the dose rates for all isotopes and exposure pathways for an assumed 1 m thick cover. In general, the dose is significantly reduced by the application of the cover. The doses from Ra-226 and Th-232 decreased to zero in the first 100 years, for K-40 except at the Ajka red mud reservoir. The dose of potassium starts to increase between 30 and 50 years. After 100 years, the contribution of K-40 decreases, while the exposure to Th-232 and Ra-226 starts to increase significantly.

A



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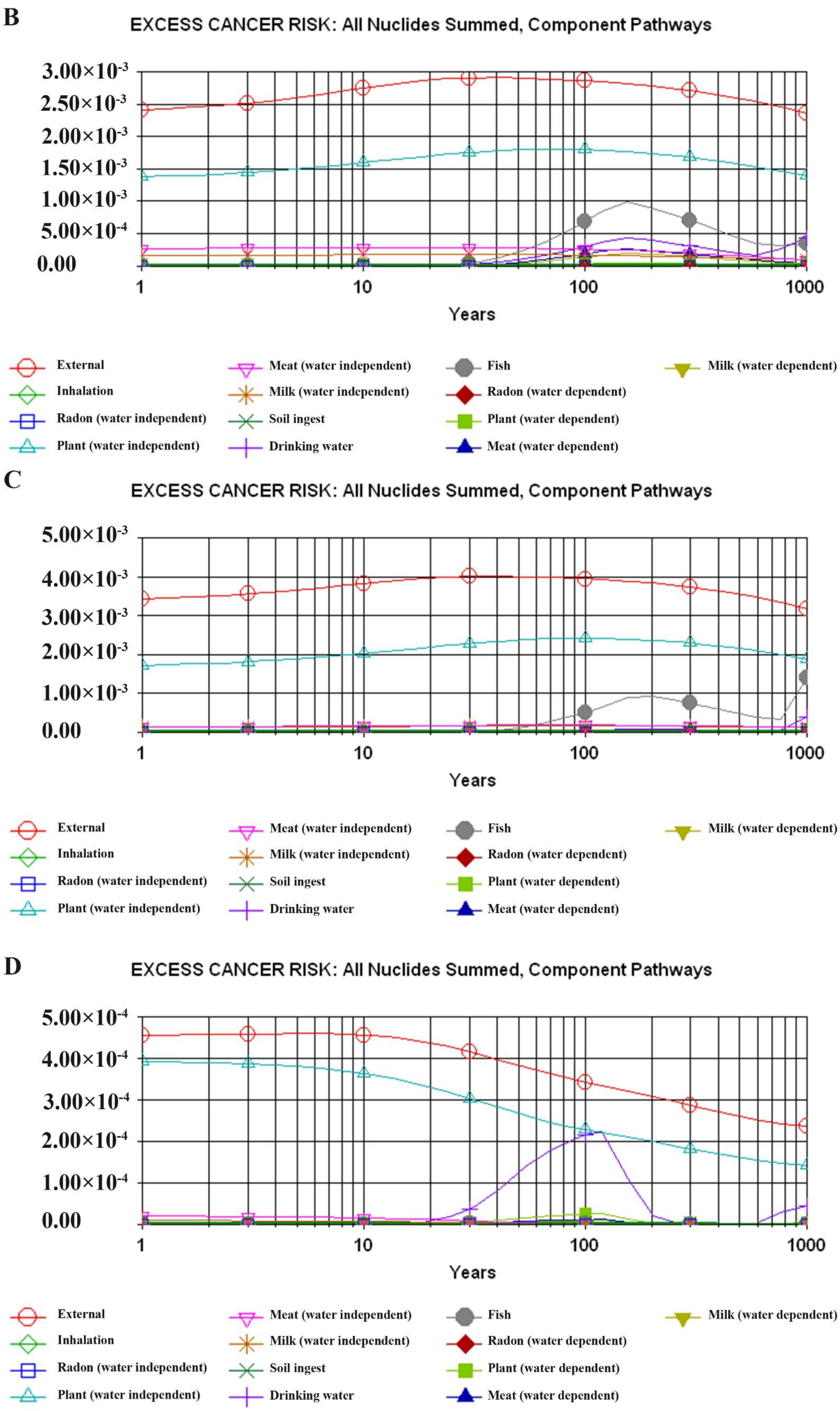

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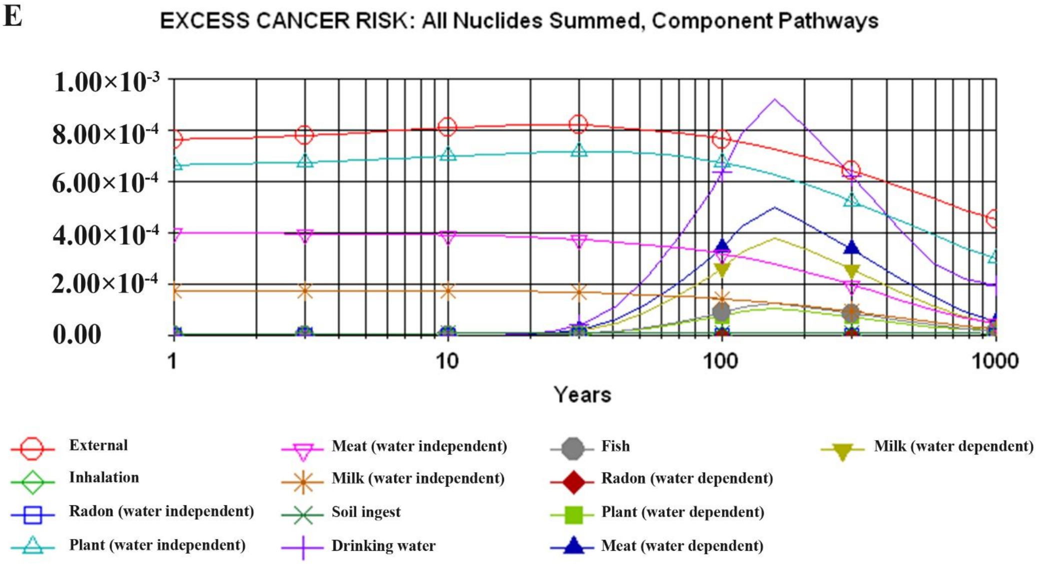
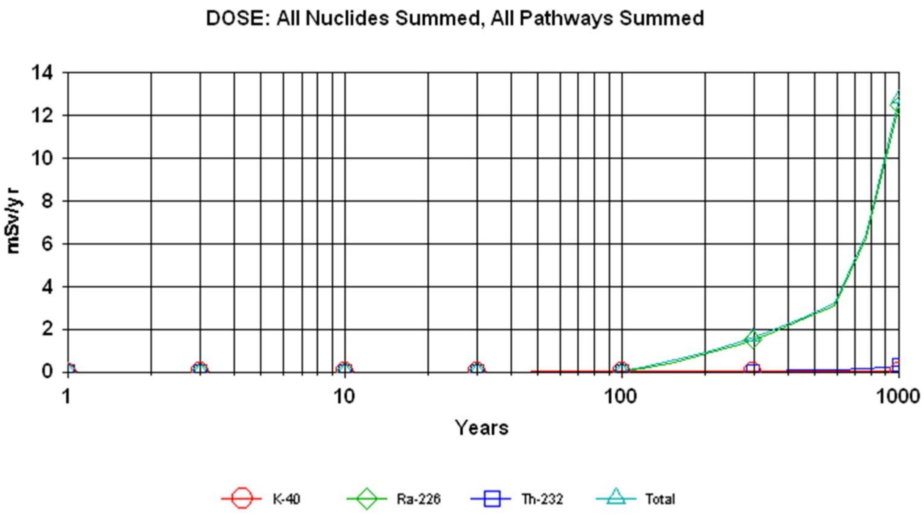


Figure 5. Contribution of the different exposure pathways to cancer risk. (**A**) Ajka coal, (**B**) Ajka red mud, (**C**) Almásfüzitő red mud, (**D**) Zalatárnok drilling mud, (**E**) Úrkút manganese.

A



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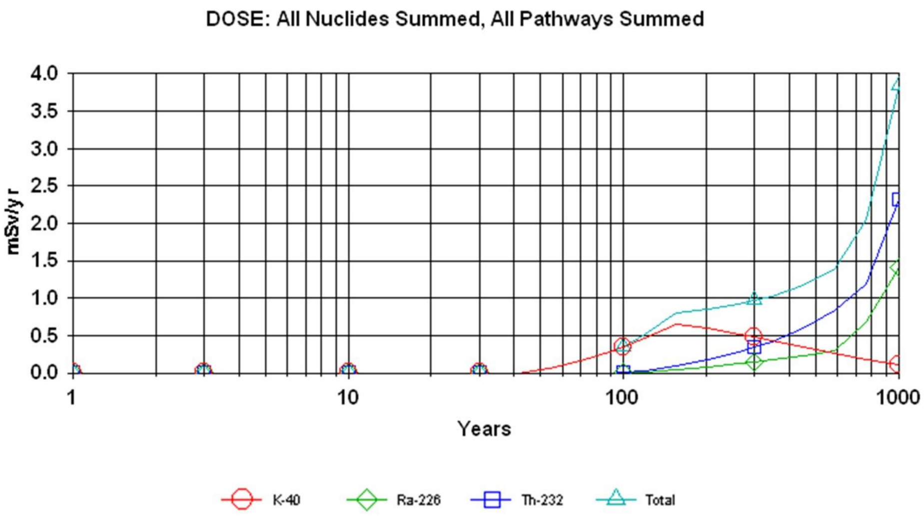

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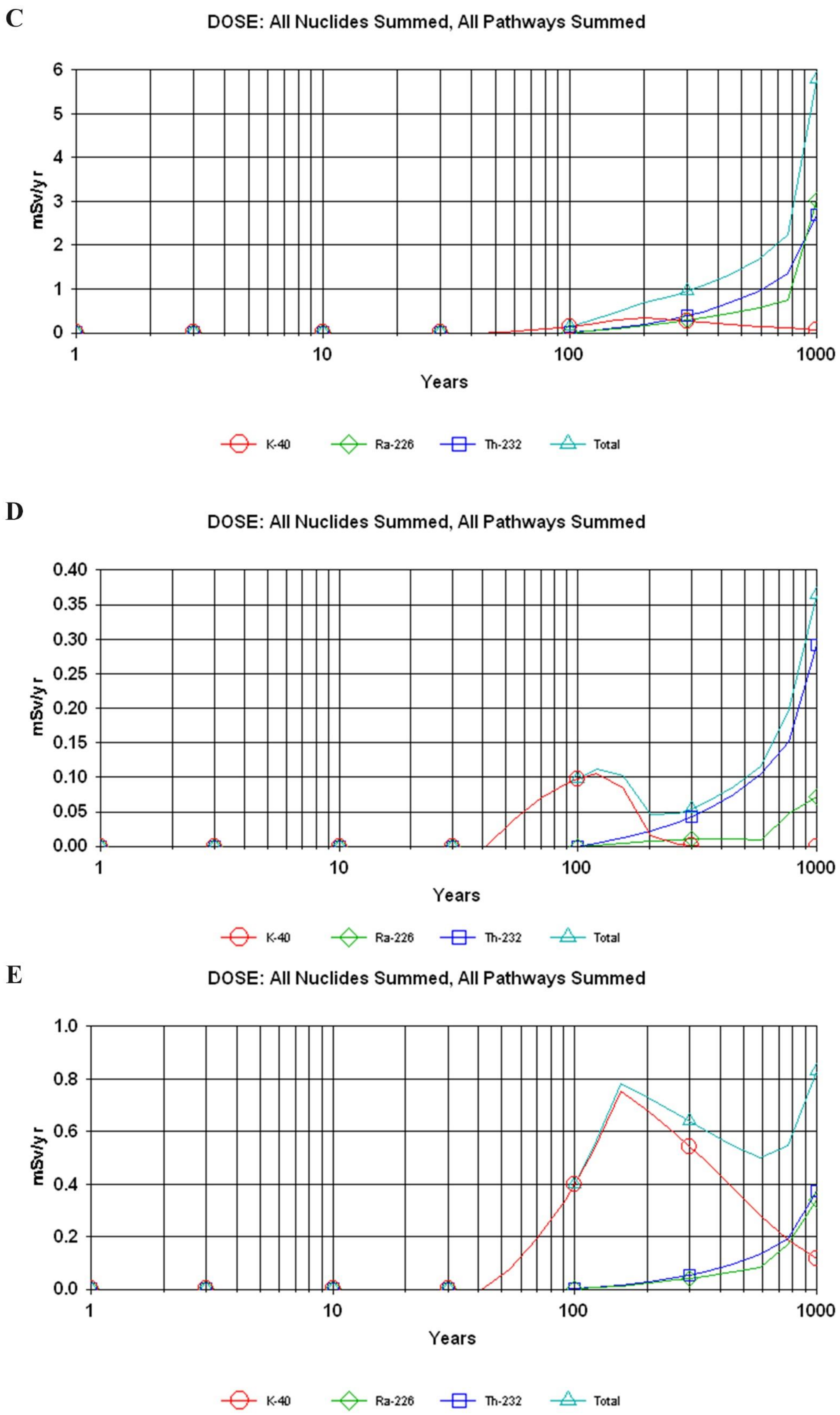

Figure 6. Total annual dose of all radionuclides using a cover (1 m). (**A**) Ajka coal, (**B**) Ajka red mud, (**C**) Almásfüzitő red mud, (**D**) Zalatárnok drilling mud, (**E**) Úrkút manganese.

RESRAD-BIOTA

RESRAD-BIOTA code was used to estimate the radiation exposure of terrestrial organisms (plants and animals). Table 4 shows the values of the sum ratio factor, biota concentration guide (BCG), external dose, internal dose, and total dose of the investigated radionuclide in terrestrial plants by soil media.

Table 4. The sum ratio factor, biota concentration guide (BCG), external dose, internal dose, and total dose of the investigated radionuclide in terrestrial plant by soil media.

Risk Parameter	Radionuclides	Coal Ash	Red Mud (Ajka)	Red Mud (Almásfüzitő)	Drilling Mud	Manganese Residue
Sum ratio factor $\left(\frac{dose\ in\ biota}{dose\ limit}\right)$	Ra-226	1.88×10^{-1}	1.71×10^{-2}	3.26×10^{-2}	2.91×10^{-3}	4.89×10^{-3}
	Th-232	3.77×10^{-5}	2.80×10^{-4}	3.23×10^{-4}	4.00×10^{-5}	4.57×10^{-5}
	K-40	1.10×10^{-3}	5.56×10^{-3}	9.40×10^{-4}	9.84×10^{-3}	1.19×10^{-2}
BCG [Bq/kg]	Ra-226	1.06×10^{4}	1.06×10^{4}	1.06×10^{4}	1.06×10^{4}	1.06×10^{4}
	Th-232	8.75×10^{5}	8.75×10^{5}	8.75×10^{5}	8.75×10^{5}	8.75×10^{5}
	K-40	5.10×10^{4}	5.10×10^{4}	5.10×10^{4}	5.10×10^{4}	5.10×10^{4}
External dose [Gy/d]	Ra-226	7.67×10^{-5}	6.98×10^{-6}	1.33×10^{-5}	1.19×10^{-6}	1.99×10^{-6}
	Th-232	5.52×10^{-9}	4.10×10^{-8}	4.74×10^{-8}	5.86×10^{-9}	6.70×10^{-9}
	K-40	5.22×10^{-7}	2.65×10^{-6}	4.45×10^{-7}	4.68×10^{-6}	5.66×10^{-6}
Internal dose [Gy/d]	Ra-226	1.80×10^{-3}	1.64×10^{-4}	3.13×10^{-4}	2.79×10^{-5}	4.69×10^{-5}
	Th-232	3.71×10^{-7}	2.76×10^{-6}	3.19×10^{-6}	3.94×10^{-7}	4.50×10^{-7}
	K-40	1.04×10^{-5}	5.30×10^{-5}	8.96×10^{-6}	9.37×10^{-5}	1.13×10^{-4}
Total dose [Gy/d]	Ra-226	1.88×10^{-3}	1.71×10^{-4}	3.26×10^{-4}	2.91×10^{-5}	4.89×10^{-5}
	Th-232	3.77×10^{-7}	2.80×10^{-6}	3.23×10^{-6}	4.00×10^{-7}	4.57×10^{-7}
	K-40	1.10×10^{-5}	5.56×10^{-5}	9.40×10^{-6}	9.84×10^{-5}	1.19×10^{-4}

The BCG levels in soil were 1.06×10^4 Bq/kg for Ra-226, 8.75×10^5 Bq/kg for Th-232, and 5.10×10^4 Bq/kg for K-40 for the terrestrial plants. BCG values indicate the radionuclide concentration at which the recommended dose limit is exceeded. As no concentration higher than the BCG value is observed in any of investigated material, no dose limit is exceeded.

SRF values ranged from 2.91×10^{-3} to 1.88×10^{-1} for Ra-226, from 3.77×10^{-5} to 3.23×10^{-4} for Th-232, and from 9.40×10^{-4} to 1.19×10^{-2} for K-40. In all cases the criterion that the SRF < 1 is met.

The external dose rate of terrestrial plants due to the exposure to Ra-226, Th-232, and K-40 is in the range of 1.19×10^{-6} Gy/d and 1.33×10^{-5} Gy/d; 5.52×10^{-9} Gy/d and 4.74×10^{-8} Gy/d; 4.45×10^{-7} Gy/d and 5.66×10^{-6} Gy/d, respectively.

The internal dose values from the radionuclides in the soil ranged from 2.79×10^{-5} Gy/d to 1.80×10^{-3} for Ra-226, from 3.71×10^{-7} to 3.19×10^{-6} for Th-232, and from 8.96×10^{-6} to 1.13×10^{-4} for K-40. All dose rate values are below the recommended dose limit, which is 0.01 Gy/d for plants.

Considering the external and internal dose from all radionuclides, the highest values were determined for coal (1.89×10^{-3}), and the lowest for drilling mud (1.28×10^{-4}).

For terrestrial animals, the calculated values are given in Table 5 for an average weight of 150 kg, and in Table 6 for an average weight of 800 kg. In general, the difference between the values calculated for the two body masses is less than 10% for Ra-226 and Th-232, and between 15% and 22% for K-40.

Table 5. Risk parameter in terms of radionuclides and materials (150 kg).

Risk Parameter	Radionuclides	Coal Ash	Red Mud (Ajka)	Red Mud (Almásfüzitő)	Drilling Mud	Manganese Residue
Sum ratio factor $\left(\frac{dose\ in\ biota}{dose\ limit}\right)$	Ra-226	7.91×10^{-1}	7.20×10^{-2}	1.37×10^{-1}	1.23×10^{-2}	2.06×10^{-2}
	Th-232	2.28×10^{-4}	1.69×10^{-3}	1.95×10^{-3}	2.42×10^{-4}	2.76×10^{-4}
	K-40	4.41×10^{-3}	2.24×10^{-2}	3.78×10^{-3}	3.96×10^{-2}	4.78×10^{-2}
BCG [Bq/kg]	Ra-226	2.53×10^{3}	2.53×10^{3}	2.53×10^{3}	2.53×10^{3}	2.53×10^{3}
	Th-232	1.45×10^{5}	1.45×10^{5}	1.45×10^{5}	1.45×10^{5}	1.45×10^{5}
	K-40	1.27×10^{4}	1.27×10^{4}	1.27×10^{4}	1.27×10^{4}	1.27×10^{4}
External dose [Gy/d]	Ra-226	7.67×10^{-5}	6.98×10^{-6}	1.33×10^{-5}	1.19×10^{-6}	1.99×10^{-6}
	Th-232	5.52×10^{-9}	4.10×10^{-8}	4.74×10^{-8}	5.86×10^{-9}	6.70×10^{-9}
	K-40	5.22×10^{-7}	2.65×10^{-6}	4.45×10^{-7}	4.68×10^{-6}	5.66×10^{-6}
Internal dose [Gy/d]	Ra-226	7.14×10^{-4}	6.50×10^{-5}	1.24×10^{-4}	1.11×10^{-5}	1.86×10^{-5}
	Th-232	2.22×10^{-7}	1.65×10^{-6}	1.91×10^{-6}	2.36×10^{-7}	2.70×10^{-7}
	K-40	3.89×10^{-6}	1.97×10^{-5}	3.34×10^{-6}	3.49×10^{-5}	4.22×10^{-5}
Total dose [Gy/d]	Ra-226	7.91×10^{-4}	7.20×10^{-5}	1.37×10^{-4}	1.23×10^{-5}	2.06×10^{-5}
	Th-232	2.28×10^{-7}	1.69×10^{-6}	1.95×10^{-6}	2.42×10^{-7}	2.76×10^{-7}
	K-40	4.41×10^{-6}	2.24×10^{-5}	3.78×10^{-6}	3.96×10^{-5}	4.78×10^{-5}

Table 6. Risk parameter in terms of radionuclides and materials (800 kg).

Risk Parameter	Radionuclides	Coal Ash	Red Mud (Ajka)	Red Mud (Almásfüzitő)	Drilling Mud	Manganese Residue
Sum ratio factor $\left(\frac{dose\ in\ biota}{dose\ limit}\right)$	Ra-226	7.91×10^{-1}	7.20×10^{-2}	1.37×10^{-1}	1.23×10^{-2}	2.06×10^{-2}
	Th-232	2.49×10^{-4}	1.85×10^{-3}	2.13×10^{-3}	2.64×10^{-4}	3.02×10^{-4}
	K-40	3.71×10^{-3}	1.88×10^{-2}	3.18×10^{-3}	3.32×10^{-2}	4.02×10^{-2}
BCG [Bq/kg]	Ra-226	2.53×10^{3}	2.53×10^{3}	2.53×10^{3}	2.53×10^{3}	2.53×10^{3}
	Th-232	1.33×10^{5}	1.33×10^{5}	1.33×10^{5}	1.33×10^{5}	1.33×10^{5}
	K-40	1.51×10^{4}	1.51×10^{4}	1.51×10^{4}	1.51×10^{4}	1.51×10^{4}
External dose [Gy/d]	Ra-226	7.67×10^{-5}	6.98×10^{-6}	1.33×10^{-5}	1.19×10^{-6}	1.99×10^{-6}
	Th-232	5.52×10^{-9}	4.10×10^{-8}	4.74×10^{-8}	5.86×10^{-9}	6.70×10^{-9}
	K-40	5.22×10^{-7}	2.65×10^{-6}	4.45×10^{-7}	4.68×10^{-6}	5.66×10^{-6}
Internal dose [Gy/d]	Ra-226	7.14×10^{-4}	6.50×10^{-5}	1.24×10^{-4}	1.11×10^{-5}	1.86×10^{-5}
	Th-232	2.43×10^{-7}	1.81×10^{-6}	2.09×10^{-6}	2.58×10^{-7}	2.95×10^{-7}
	K-40	3.18×10^{-6}	1.61×10^{-5}	2.73×10^{-6}	2.85×10^{-5}	3.45×10^{-5}
Total dose [Gy/d]	Ra-226	7.91×10^{-4}	7.20×10^{-5}	1.37×10^{-4}	1.23×10^{-5}	2.06×10^{-5}
	Th-232	2.49×10^{-7}	1.81×10^{-6}	2.13×10^{-6}	2.64×10^{-7}	3.02×10^{-7}
	K-40	3.71×10^{-6}	1.61×10^{-5}	3.18×10^{-6}	3.32×10^{-5}	4.02×10^{-5}

The BCG levels in soil were 2.53×10^{3} Bq/kg for Ra-226, 1.33×10^{5} Bq/kg for Th-232, and 1.51×10^{4} Bq/kg for K-40. BCG values indicate the radionuclide concentration at which the recommended dose limit is exceeded. As no concentration higher than the BCG value can be measured in any of investigated material, no dose limit is exceeded.

SRF values ranged from 1.23×10^{-2} to 7.91×10^{-1} for Ra-226, from 2.49×10^{-4} to 2.13×10^{-3} for Th-232, and from 3.18×10^{-3} to 4.02×10^{-2} for K-40. In all cases, the criterion that the SRF is less than 1 is met.

The external dose rate of terrestrial animals due to the exposure to Ra-226, Th-232, and K-40 is in the range of 1.19×10^{-6} Gy/d and 1.33×10^{-5} Gy/d; 5.52×10^{-9} Gy/d and 4.74×10^{-8} Gy/d; 4.45×10^{-7} Gy/d and 5.66×10^{-6} Gy/d, respectively.

The internal dose values from the radionuclides in the soil ranged from 1.11×10^{-9} Gy/d to 7.14×10^{-4} for Ra-226, from 2.22×10^{-7} to 2.09×10^{-6} for Th-232, and from 2.73×10^{-6} to 4.22×10^{-5} for K-40. All dose rate values are below the recommended dose limit, which is 0.01 Gy/d for plants.

Considering the external and internal dose from all radionuclides, the highest values were determined for coal (7.96×10^{-4} Gy/d) in the case of 150 kg average mass and the lowest for drilling mud (4.58×10^{-5} Gy/d) in the case of 800 kg average mass.

4. Conclusions

The main treatment option for industrial by-products from human activities is disposal in the environment. Some of them contain elevated levels of radionuclides, which can present a serious long-term radiological risk to biota and humans. Biota, as an environmental element, and its components are in a complex and complicated interaction with other elements of the environmental system, so any intervention can directly or indirectly affect its state.

In this study, five NORM environmental repositories in Hungary have been investigated using the RESRAD-ONSITE and RESRAD-BIOTA codes. RESRAD-ONSITE was applied to estimate the cancer risk from the natural radionuclides in the storage. RESRAD-BIOTA code is for estimating radiation dose to nonhuman, terrestrial biota.

RESRAD-ONSITE code was used for dose and risk estimation in the work of Njinga and Tshivhase. They investigated the Tudor Shaft Mine Tailing Sites (South Africa) [33]. Their maximum calculated dose was 1.639 mSv/year, which is similar to the Úrkút manganese residue. In our study, the maximum total dose was ten times higher, 12.38 mSv/year on the Ajka coal ash depository.

Bello et al. [34,35] carried out modelling in a granite-based field in Nigeria. In their study, the total dose was 1.59 mSv/year, which is similar to the measurement results from Africa.

With the RESRAD-BIOTA, the BCG level, SRF, and internal and external doses are calculated. The BCG levels in soil were 1.06×10^4 Bq/kg (plant) and 2.53×10^3 Bq/kg (animal) for Ra-226, 8.75×10^5 Bq/kg (plant) 1.33×10^5 Bq/kg (animal) for Th-232 and 5.10×10^4 Bq/kg and 1.51×10^4 Bq/kg (animal) for K-40. The values of the dose rates determined with RESRAD-BIOTA version 1.8 are below the recommended dose limit of DOE.

Cujic and Dragovic carried out a study in the area surrounding the largest Serbian coal-fired power plant. The highest total dose rate was 3.45×10^{-4} Gy/d, which is similar to our estimated dose in the case of the coal ash depository.

In Hungary, the dose limit for the public from terrestrial radionuclides is 1 mSv/year, which has been exceeded in all cases except for drilling mud (Zalatárnok). By applying a one-meter-thick cover, the dose is estimated to be below the limit value.

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