Comparative Assessment of Radiation Background Due to Natural and Artificial Radionuclides in Soil in Specific Areas on the Territories of State of Washington (USA) and Lithuania

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Abstract Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in soil samples collected at Mount Rainier National Park and Satsop Nuclear Power Plant zone in State of Washington (USA) and Curonian Spit National Park and Ignalina Nuclear Power Plant zone in Lithuania have been measured by gamma spectrometry. The results have been compared with the worldwide measured average values reported by the UNSCEAR. Higher activity concentrations of ^{137}Cs were found in Curonian Spit National Park due to Chernobyl accident fallout. The radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), the internal hazard index (H_{in}), the absorbed dose rate (D), and the annual effective dose rate ($AEDR$) were also calculated and compared with the international recommended values. These calculated hazard indices used to estimate the potential radiological health risk in soil and the dose rate associated with it are below their permissible limit.

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1 Introduction

The background radiation in the environment is emitted by natural and anthropogenic radiation sources. The estimations show that almost 80% of the collective radiation exposure of the world's population is generated by natural sources; i.e., natural radiation derives from the primordial radionuclides found in the Earth's crust and the cosmogenic radionuclides produced from the interactions of cosmic-ray occurring in the atmosphere (UNSCEAR 2008).

One of major sources of radiation exposure to a population is soil, which allows the transfer of radionuclides into the environment. The patterns of gamma emitters ^{238}U, ^{232}Th, and ^{40}K in soil significantly depend on the radionuclide distribution in rocks from which they originate and on the processes through which the soils are concentrated (Ahmad et al. 2015; Durusoy and Yildirim 2017). Higher radiation levels are generally more specific to igneous rocks, such as granite, and lower levels to sedimentary rocks. The natural environmental radiation is mostly determined by geological and geographical conditions (Song et al. 2012; Baykara and Dogru 2009; Ravisankar et al. 2012; Nguelem et al. 2016; Alshahri and El-Taher 2019).

It is important to know the distribution pattern of both anthropogenic and natural radionuclides in order to assess the radiation exposure to the population. Isotopic determination of natural and anthropogenic radionuclides is needed for nuclear safeguards, environmental monitoring, and nuclear forensic studies. The nuclear weapon tests and accidents at nuclear reactors and

discharges of radioactive waste from nuclear installations are the main sources of anthropogenic radionuclides in the environment. ^{137}Cs is the most important constituent detected by its gamma radiation in the environment among the artificial isotopes released as decay products, such as ^{90}Sr and ^{95}Zr (Turhan et al. 2012; Abdi et al. 2009). This fission product is important due to its long half-life of 30.17 years, and because it is a high energetic gamma emitter (662 keV) contributing to people's dose due to the external radiation exposure (Bara et al. 2012; Eke et al. 2015). This radionuclide could be produced as the results of nuclear tests, normal function of nuclear power plants, or reactor accidents that significantly contribute to soil contamination because ^{137}Cs could be found in superficial layers of soil and be absorbed by plant roots conducting that to the human food chain (Changizi et al. 2013). The risk from ^{137}Cs highly depends on its diffusion rates in soil. Slow migration of ^{137}Cs in soil results in higher internal irradiation due to higher absorption by plants roots especially from the top surface of 5 cm depth (Butkus and Konstantinova 2008).

The northern hemisphere is more contaminated than the southern one due to the presence of large quantities of radioactive isotopes (especially ^{14}C, ^{137}Cs, and ^{90}Sr) released into the atmosphere during nuclear weapons tests (UNSCEAR 2000). Higher radioactivity of the northern hemisphere is also due to other factors, such as nuclear power plant accidents. The most representative examples are the accidents at Chernobyl (1986) and Fukushima Daiichi (2011), which resulted in the release of large amounts of radionuclides into the atmosphere.

The nuclear power accident, which had occurred in Chernobyl in 1986, had negative consequences for the radioactive contamination in the territory of Lithuania (Šidiškienė 2001; Nedveckaitė 2004). The radioactive dust cloud, which had formed in the atmosphere after the accident, passed through the south-western part of the country (Fig. 1). After this accident, the activity density of ^{137}Cs deposition in Lithuania reached 18.5 kBq m^{-2}. Maximum levels of radioactive deposition have been found in the western and south-western districts. The maximum activity has been recorded in the soil of Curonian Spit; i.e., approximately 19.7 kBq m^{-2}, in other southern and a south-western part of the country, the maximum activity was approximately 1.6 kBq m^{-2} (Butkus et al. 2001; Lubytė and Antanaitis 2004). ^{137}Cs concentrations in southern, south-western, and western Lithuania were quite different, i.e., varied

from 2 to 439 Bq kg^{-1}. Radioactive strontium and plutonium isotopes resulting from the reactor where the accident had occurred were released later when the temperature of the heated reactor reached thousand degrees, and air masses moved towards the north, east, and later south; therefore, the contamination of the Lithuanian soil with these radionuclides had varied insignificantly (Nedveckaitė 2004).

It has been noticed the environment is cleaned through radioactive decay only after 10 half-life times of degradation. Chernobyl disaster emissions also contained significant quantity of long-lived radionuclides; thus, their contamination is long lasting. The nature of contamination varies over time, radionuclides migrate vertically and horizontally, and they are raised by wind above the ground and deposit on other regions and are eliminated with plants, etc. (Butkus 2006).

It is well known that ^{137}Cs slowly migrates to the deeper soil layers, and the vertical migration rate is about 0.1–0.2 cm year^{-1} (Butkus and Konstantinova 2008). Therefore, even now, radioactive cesium is found to be in the upper layers of soil where it is absorbed by plants and can be ingested by humans through the food chain and increase contamination in this way.

The main objective of this study is to determine the activity concentrations and radiological hazard parameters of natural and artificial radionuclides (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) in the soil at the unique and specific areas from the sites of two different countries, i.e., Lithuania and USA (State of Washington).

2 Materials and Methods

2.1 Measuring Location

The research was carried out in territories of State of Washington and Lithuania. Two unique areas have been chosen for the research: recreation zone (National Park) and the nuclear power plant zone.

Lithuania is located in central part of Europe on the south-eastern coast of the Baltic Sea. The territories of Curonian Spit National Park and Ignalina Nuclear Power Plant have been chosen for the research in Lithuania (Fig. 2).

Curonian Spit National Park is a recreation zone in the Curonian Spit, which is included on the UNESCO World Heritage List. Curonian Spit National Park is characterized by a very clean environment and a

Fig. 1 Radioactive cloud trajectory after the accident occurred in Chernobyl Nuclear Power Plant on April 26, 1986
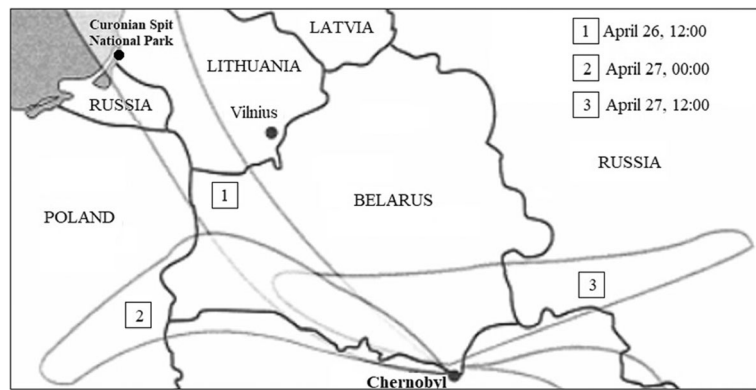
minimal urbanization. The Curonian Spit is a unique and vulnerable, sandy, and wooded cultural landscape on a coastal spit, which features small Curonian lagoon settlements. Currently, forests and sands dominate in the Curonian Spit. Urbanized areas cover only about 6% of the land. The condition of the beach depends on the geological structure of the underwater shore slope, the amount of sediment, and the lithodynamic situation. The coast is dominated by fine sand. A part of the coast is covered with stones. The soil samples were taken from recreation zone (beach) and urbanized part of Curonian Spit National Park.

Ignalina district is special because of the nuclear power plant (which is currently being decommissioned). It is the only nuclear power plant in the Baltic States. The Ignalina Nuclear Power Plant contained two Soviet-designed RBMK-1500 water-cooled graphite-moderated channel-type power reactors. After the Chernobyl disaster occurred in April of 1986, the reactor was de-rated to 1360 MW. Each unit of the power plant was equipped with two K-750-65/3000 turbines with 800 MW generators. The samples in Ignalina were taken from the sites, which are located next to the Ignalina Nuclear Power Plant. The whole territory is covered by the refilled technogenic soil. The sediment-subsidized depth at the whole site varies from 0.2 to 10.0 m. The maximum thickness of the soil is in the central and western parts of the site where it exceeds 4.0–10.0 m. The investigated refill soil covering the site is composed of sandy loam, silt loam, peat, and sand of different coarseness with the layer of peat and black-earth. The soil in the layers deeper than 1 m was mixed with building rubble, namely, concrete waste, reinforcement, plastics, aggregate, and timber. In some places there is an underlaying sandy loam below the above mentioned soil.

The territories of Satsop, Washington, and Mount Rainier National Park have been chosen for the research in the USA (Fig. 3).

Satsop Nuclear Power Plant is located on 650 ha on the Satsop Site near Elma in Grays Harbor County, Washington. Satsop Nuclear Power Plant is known as Washington Nuclear Project Nos. 3 and 5, abbreviated as WNP-3 and WNP-5. WNP-3 and WNP-5 were two of five nuclear power plants on which construction was started by the Washington Public Power Supply System in order to meet projected electricity demand in the Pacific Northwest. The construction of WNP-3 and WNP-5 were halted and only WNP-2 was completed and put into operation. Twenty-five samples were taken from different areas of Satsop Nuclear Power Plant.

Mount Rainier National Park is an American National Park located in south-east Pierce County and northeast Lewis County in State of Washington. Mount Rainier is a large active stratovolcano in Cascadia located south-south-east of Seattle. It is the highest mountain in the US state of Washington, and of the Cascade Range of the Pacific Northwest, the most topographically prominent mountain in the contiguous United States and the tallest in the Cascade Volcanic Arc. Wildlife abounds in the park's ecosystems. The soil samples were taken from different areas of Mount Rainier National Park. The soil samples were taken from recreation zone and urbanized part of Mount Rainier National Park.

2.2 Measurement Methodology

Samples were placed in standard geometry plastic containers. The coarse particles were mechanically comminuted to homogeneously fill the containers.

Fig. 2 Sampling location in Lithuania

Fig. 3 Sampling location in State of Washington

The specific activities of natural and artificial radionuclides ^{232}Th, ^{226}Ra, ^{40}K, and ^{137}Cs were determined experimentally in the samples of the dominant and technogenic soil from Lithuania (Curonian Spit National Park and Ignalina Nuclear Power Plant zone) and State of Washington (Satsop Nuclear Power Plant zone and Mount Rainier National Park). The samples were crushed, dried at 105 °C up to constant weight, and sealed in the plastic measuring containers of 200 ml capacity for 4 weeks for balance between ^{226}Ra and ^{232}Th and stabilization of their decay products.

The samples were analyzed by using a CANBERRA γ-spectrometric system with an HPGe detector (model GC2520, with 26.2% relative efficiency, 1.76 keV/ 1.33 MeV resolution according to the gamma line of ^{137}Cs at 661.62 keV). The measurements were carried out in standard geometry (frustum) containers and at the known efficiencies (1.8–3.3%) depending on the density of samples. The calibration quality was certified by using the IAEA standards. The measurement errors of ^{137}Cs activity concentration in the samples were evaluated by using the GENIE software program.

The samples were tested at least for 90.000 s. The radionuclides were identified by the following energies: 186 keV of ^{226}Ra, 295 keV and 352 of ^{214}Pb (^{226}Ra progeny), 583 keV of ^{208}Tl (^{232}Th progeny), and 1460 keV of ^{40}K.

3 Results and Discussions

3.1 Activity Concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in Soil Samples

The activity concentrations (Bq kg^{-1}) of the naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K and the artificially occurring radionuclide ^{137}Cs were determined in the soil samples from Lithuania and State of Washington.

The activity concentration data with minimum, maximum, mean, median, and standard deviation are presented in Table 1. The ^{40}K activity concentration mean values range from 124.6 Bq kg^{-1} in Satsop NPP area to 470.1 Bq kg^{-1} in Ignalina NPP area, mean values of ^{232}Th activity concentration vary to a lesser extent (from 3.1 Bq kg^{-1} in Mount Rainier National Park to 6.6 Bq kg^{-1} in Curonian Spit National Park), and the ^{226}Ra activity concentration mean maximum was

observed in Mount Rainier National Park (22.9 Bq kg^{-1}), and minimum is in Ignalina NPP area (13.7 Bq kg^{-1}).

The radionuclide activity concentration values varied in different areas depending on their geological structures, rainfall amounts, and elevations. Moreover, the composition of the soil and the content of fertilizers in the soil are also important variables. In sandy soils, a large amount of ^{40}K can be observed, while in light, organic soils, the activity concentrations of ^{40}K and ^{226}Ra as well as ^{232}Th are low. However, the natural activity concentration values in all soil samples were less than the global limit average (median values 30 Bq kg^{-1} for ^{232}Th, 35 Bq kg^{-1} for ^{226}Ra, and 400 Bq kg^{-1} for ^{40}K) (UNSCEAR 2000).

As shown in Table 1, the values of the activity concentration of ^{137}Cs in Washington Nuclear Project range from 0.43 to 11.61 Bq kg^{-1}, averaging 4.2 Bq kg^{-1}, while the values in Mount Rainier range from 2.98 to 33.21 Bq kg^{-1}, averaging 13.1 Bq kg^{-1}; beside, in both areas, the samples with a value below the detection limit were observed. Furthermore, in both areas, the increased values of ^{137}Cs activity (11.61 and 33.21 Bq kg^{-1}) were detected.

As is well known, ^{137}Cs in the US territory in the main is originated from nuclear weapon tests in the 1950s–1960s (bomb-derived ^{137}Cs) (Tompson et al. 2002), and to some extent from the nuclear accident as Fukushima Daiichi NPP accident, whatever in Lithuania territory this radionuclide has both bomb-derived and Chernobyl accident. In April of 1986, Lithuania, especially the western part, was directly in the path of air masses coming from Chernobyl (Fig. 1). The activity concentration of this radionuclide in Lithuania ranges from negligible values to very significant one (up to 33.6 Bq kg^{-1} in forest soils of the Curonian Spit) as the trace of the Chernobyl fallout. The ^{137}Cs activity concentration values in Ignalina NPP area (north-eastern part of Lithuania) are less, and fluctuate to a lesser extent, from 4.8 to 8.4 Bq kg^{-1}. The higher values of ^{137}Cs activity concentration in State of Washington (33.21 Bq kg^{-1} in Mount Rainier National Park) can be affected by nuclear weapon tests.

3.2 Radiation Hazard Indices

It is very important to conduct measurements of natural radiation and artificial radiation due to accident-induced ^{137}Cs for the purpose of monitoring the region for

Table 1 Descriptive statistics of radionuclide activity concentration, Bq kg^{-1}: mean value (M.V.), standard deviation (σ), and range (in brackets)

Location	^{40}K M.V. $\pm \sigma$ (range)	^{232}Th M.V. $\pm \sigma$ (range)	^{226}Ra M.V. $\pm \sigma$ (range)	^{137}Cs M.V. $\pm \sigma$ (range)
Lithuania				
Curonian Spit National Park (sand)	321.0 ± 82.1 (150.3÷578.3)	5.3 ± 1.0 (1.0÷19.8)	14.7 ± 2.03 (2.8÷29.1)	9.4 ± 3.2 (0.9÷20.7)
Curonian Spit National Park (forest soil)	253.1 ± 58.9 (140.8÷502.6)	6.6 ± 1.5 (3.2÷24.1)	15.1 ± 3.7 (1.5÷31.4)	23.2 ± 3.7 (1.1÷33.6)
Ignalina Nuclear Power Plant	470.1 ± 93.0 (185.2÷620.7)	4.5 ± 0.8 (0.7÷21.3)	13.7 ± 3.1 (4.3÷37.4)	7.2 ± 2.1 (4.8÷8.4)
USA Washington St.				
Satsop Nuclear Power Plant	124.6 ± 52.2 (22.3÷181.0)	3.3 ± 1.5 (1.92÷6.02)	18.5 ± 9.8 (2.73÷31.43)	4.2 ± 1.4 (0.43÷11.61)
Mount Rainier National Park	157.5 ± 27.1 (125.0÷185.7)	3.1 ± 0.7 (2.39÷3.76)	22.9 ± 3.6 (19.13÷27.44)	13.1 ± 7.4 (2.98÷33.21)

human health. For estimation of risk due to that, various dosimetric parameters are used.

The radiation risk due to natural radionuclides in soil can be calculated by using various radiation hazard indices. The reference value for each radionuclide is an acceptable effective dose equivalent.

A hazard index represents a ratio with the numerator being a determined value for a particular contaminant and the denominator a reference value for that contaminant. The reference value represents an amount that is associated with an acceptable level of risk. If the hazard index's value is bigger than unity, it requires some sort of remediation.

The external hazard index formula represents the risk associated with the external source of gamma rays. In soil, the external hazard index is calculated by using the following formula:

$$H_{ex} = \frac{A_{226_{Ra}}}{370} + \frac{A_{232_{Th}}}{259} + \frac{A_{40_K}}{4810} \qquad (1)$$

where $A_{226_{Ra}}$, $A_{232_{Th}}$, and A_{40_K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bq kg^{-1}. If the value of H_{ex} exceeds unity, it is concluded that the potential external dose to exposed persons exceeds the acceptable level (Ali et al. 2013a).

The internal hazard index is defined as follows:

$$H_{in} = \frac{A_{226_{Ra}}}{185} + \frac{A_{232_{Th}}}{259} + \frac{A_{40_K}}{4810} \qquad (2)$$

where $A_{226_{Ra}}$, $A_{232_{Th}}$, and A_{40_K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bq kg^{-1}, and the internal radiation hazard index (H_{in}) should also be less than unity to avoid radiation hazards to the respiratory organs (Ali et al. 2013b).

In this study, the hazard indices Ra$_{eq}$, H_{ex}, and H_{in} were calculated and provided in Table 2. The external hazard index H_{ex} represents the risk associated with the external source of gamma rays. A sample is considered safe if H_{ex} is ≤ 1. The results show that all samples H_{ex} is less than 1, and it means that all samples are safe to handle. H_{in} should also be less or equal to unity to avoid radiation hazards to the respiratory organs. It was found that H_{ex} was less in both countries than unity H_{in} and varied from 0.09 to 0.15.

The influence of all natural radionuclides on the potential radiation has been assessed by calculating of

Table 2 Radiation hazard indices from natural radionuclides

Location	Ra$_{eq}$	H_{ex}	H_{in}
Lithuania			
Curonian Spit National Park (sand)	24.75	0.13	0.17
Curonian Spit National Park (forest soil)	26.48	0.12	0.16
Ignalina Nuclear Power Plant	23.75	0.15	0.19
USA Washington St.			
Satsop Nuclear Power Plant	24.18	0.09	0.14
Mount Rainier National Park	28.54	0.11	0.17
Limiting value	370	1	1

the equivalent activity of radium Ra$_{eq}$. This parameter is used for assessing building materials and soil from the point of view of radiation protection (Singh et al. 2005). The Radium equivalent activity of the soil samples was calculated on the basis of the estimation that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K produce the same gamma dose rate. The values of Ra$_{eq}$ were calculated by using the following equation (Ali et al. 2013b):

$$Ra_{eq} = A_{226_{Ra}} + 1.432_{232_{Th}} + 0.077A_{40K} \qquad (3)$$

where $A_{226_{Ra}}$, $A_{232_{Th}}$, and A_{40_K} are the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in becquerel per kilogram.

It is evident from Table 2 that the radium equivalent activity is 370 times higher than the external hazard index and presents similar information but in quantitative units (Bq kg^{-1}). A material is considered to be potentially hazardous if its Ra$_{eq}$ exceeds 370 Bq kg^{-1}. Ra$_{eq}$ in our data varied from 23.75 to 28.54 Bq kg^{-1}. This shows that all samples have not exceeded the limit which is 370 Bq kg^{-1}.

3.3 Absorbed Dose Rate

According to the results of the spectrometric analysis of the environment surrounding a human, the contribution of the different radionuclides occurring in soil to the exposure was assessed. To assess the possible impact of natural radiation on a human, the results of the studies carried out in different locations in Lithuania and State of Washington have been used to calculate the absorbed dose rate (nanogray per hour) by applying the Monte Carlo method (UNSCEAR 2000; Ibraheem et al. 2018).

The conversation factors to transform specific activities A_{40_K}, $A_{226_{Ra}}$, $A_{232_{Th}}$ in absorbed dose rate (D) at 1 m above the ground (in nanogray per hour by becquerel per kilogram) were found by applying Monte Carlo method as follows:

$$D = 0.0417A_{40K} + 0.462A_{226_{Ra}} + 0.604A_{232Th} \qquad (4)$$

where $A_{226_{Ra}}$, $A_{232_{Th}}$, and A_{40_K} are the specific activities of ^{40}K, ^{226}Ra, and ^{232}Th in Bq kg^{-1}.

Table 3 shows that all samples had lower values than the world average air absorbed dose rates of 51 nGy h^{-1} (UNSCEAR 2000). An average value of samples is

above 45–70% lower than the world average (UNSCEAR 2008).

3.4 Annual Effective Dose Rate

An effective dose rate is the most commonly used indicator of the potential biological effects associated with exposure to ionizing radiation in humans and radiation protection. The annual effective dose rate caused by natural terrestrial radionuclides was determined by using absorbed dose rate in air estimation.

Due to natural terrestrial radionuclides deposited on earth, in order to calculate the annual effective dose rate (AEDR$_{nat}$), the absorbed dose rate in air is used in the following way (Ali et al. 2013a):

$$AEDR_{nat} = 0.69D[(1-I_{in}) + SF \cdot I_{in}] \cdot 24 \cdot 365.25$$
$$\cdot 10^{-3} \qquad (5)$$

where I_{in} (0.8) represents the occupancy factor for indoor dose, SF means the shielding factor (0.2), while 0.69 is the dose conversion factor (Sv Gy^{-1}) (Ali et al. 2013b), which allows converting the absorbed dose in air to human effective dose in adults. The remaining factors are used only for unit conversions.

The values of AEDR$_{nat}$ determined in different locations are presented in Table 3. They varied from 33.33 μSv year^{-1} (Satsop Nuclear Power Plant) to 60.67 μSv year^{-1} (Ignalina Nuclear Power plant) representing from 47 to 87% of the global average (UNSCEAR 2008).

Due to the presence of ^{137}Cs in the soil, the annual effective dose rate at 1 m height above the ground level is calculated as follows:

$$AEDR_{137_{Cs}} = 0.576 \cdot 0.662 \cdot \varphi$$
$$\cdot \frac{\mu_a(E)^{tissue}}{\rho}[(1-I_{in}) + SF \cdot I_{in}] \qquad (6)$$

$$\varphi = \frac{0.8512\,A}{2[\mu_s(E)/\rho]^{soil}} \qquad (7)$$

where 0.662 is the gamma-ray energy of ^{137}Cs in MeV, φ represents the mean gamma-ray flux, $(\mu_a(E)/\rho)^{tissue}$ means the energy-dependent mass attenuation

Table 3 Absorbed dose rate (D) in air and annual effective dose rate (AEDR) due to external sources of radiation

Location	D (nGy h^{-1})	AEDR$_{nat}$ (µSv year^{-1})	AEDR$_{137_{Cs}}$ (µSv year^{-1})
Lithuania			
Curonian Spit National Park (sand)	23.38	49.51	0.22
Curonian Spit National Park (forest soil)	21.52	45.57	0.55
Ignalina Nuclear Power plant	28.65	60.67	0.17
USA (Washington St.)			
Satsop Nuclear Power Plant	15.74	33.33	0.10
Mount Rainier National Park	19.02	40.27	0.31
World average	51	70	3

coefficient of soft tissue (0.0316 cm^2 g^{-1}), 0.8512 is the gamma-ray line abundance at 0.662 MeV, and $(\mu_s(E)/\rho)^{soil}$ is the energy-dependent mass attenuation coefficient of soil (0.0780 cm^2 g^{-1}) (Tahir et al. 2006).

The effective dose rate calculated by using ^{137}Cs is provided in Table 3. The world average effective dose rate caused by anthropogenic radionuclides occurring as a result of the Chernobyl accident is 3 µSv year^{-1} (Ali et al. 2013b).

The values of AEDR$_{137_{Cs}}$ for Curonian Spit National Park and Mount Rainier National Park are from 7 to 10% of the global average. The average value of the AEDR$_{137_{Cs}}$ near Ignalina Nuclear Power Plant due to ^{137}Cs in soil is 0.17 µSv year^{-1} representing 6% of the global average. While the AEDR$_{137_{Cs}}$ due to ^{137}Cs in soil next to the Satsop Nuclear Power Plant is 0.10 µSv year^{-1} representing 3% of the world's average.

The annual effective dose rate AEDR$_{soil}$ caused by natural terrestrial (AEDR$_{nat}$) and anthropogenic radiocesium in soil (AEDR$_{137_{Cs}}$) was calculated by using the following equation:

$$AEDR_{soil} = AEDR_{nat} + AEDR_{137_{Cs}} \qquad (8)$$

The absorbed and annual effective dose rates in different locations of Lithuania and State of Washington are shown in Fig. 4.

AEDR$_{soil}$ is defined as the effective dose rate due to natural terrestrial and anthropogenic radionuclides such as ^{137}Cs deposited on the surface of earth. Due to natural radionuclides and ^{137}Cs, AEDR$_{soil}$ values ranged from 33.43 µSv year^{-1} (Satsop Nuclear Power Plant) to 60.84 µSv year^{-1} (Ignalina Nuclear Power Plant). The total dose rate due to terrestrial radionuclides is partitioned into 99.7–99.2% (natural) and 0.3–0.8% (^{137}Cs). In this case, we do not consider the effect of cosmic radiation and radionuclides present in building

Fig. 4 Absorbed (D) and annual effective dose rates (AEDR$_{soil}$) in different locations of Lithuania and State of Washington
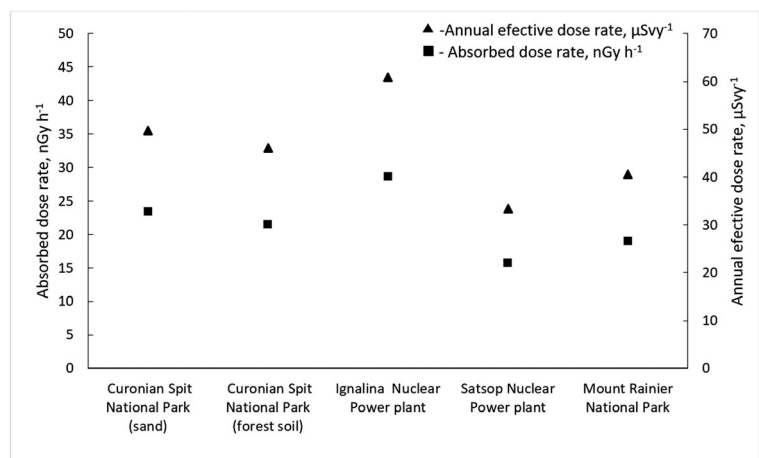
materials. Obviously, the majority of the total dose rate is determined by the radiation of natural radionuclides. It was found that artificial radionuclides are removed from the environment by radioactive decay after 10 half-life periods (Butkus 2006). Since ^{137}Cs half-life is approximately 30 years, it is obvious that we can still found this radionuclide in the soil in the territory of both State of Washington and Lithuania.

4 Conclusions

While comparing the radiation background of two countries located on different continents and in different hemispheres, it was noted that the natural radionuclides activity concentration values in all soil samples determined during this study were generally lower than the average values from all around the world; i.e., the study area radiation was within the natural limits and did not significantly differ from the radiation in other regions of the world. The variation of radionuclide activity concentration values in different areas depends on their geological structures, rainfall amounts, elevations, and mostly on the soil composition.

The higher activity concentrations of ^{137}Cs were found in few samples in State of Washington. This could have been affected by nuclear weapon tests in the 1950s–1960s. The highest values of ^{137}Cs concentration were found in Lithuania, in Curonian Spit National Park, basically in forest soil, due to Chernobyl accident fallout. The measurements showed that although ^{137}Cs activity concentrations were lower, the radionuclide still persisted in the regions.

Various dosimetric parameters, such as radiation hazard indices, absorbed dose rate, and annual effective dose rate, are used to estimate radiation risk due to radiation of natural radionuclides in soil, as well as artificial radiation due to both bomb-derived and Chernobyl or Fukushima Daiichi accident-induced ^{137}Cs.

The estimated Radium equivalent activity in all samples did not exceed limit equal to 370 Bq kg^{-1}. Absorbed dose rate of all samples also had lower values than the world average air absorbed dose rates, and the average value of samples was above 45–70% lower than the world average. The total dose rate due to terrestrial radionuclides is partitioned into 99.7–99.2% (natural) and 0.3–0.8% (^{137}Cs).

References

Abdi, M. R., Hassanzadeh, S., Kamali, M., & Raji, H. R. (2009). ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs activity concentrations along the southern coast of the Caspian Sea, Iran. *Marine Pollution Bulletin, 58*, 658–662.

Ahmad, N., Jaafar, M. S., Bakhash, M., & Rahim, M. (2015). An overview on measurements of natural radioactivity in Malaysia. *Journal of Radiation Research and Applied Science, 8*(1), 136–141.

Ali, M., Wasim, M., Iqba, S., Arif, M., & Saif, F. (2013a). Determination of the risk associated with the natural and anthropogenic radionuclides from the soil of Skardu in Central Karakoram. *Radiation Protection Dosimetry, 156*(2), 213–222.

Ali, M., Iqbal, S., Wasim, M., Arif, M., & Saif, F. (2013b). Soil radioactivity levels and radiological risk assessment in the highlands of Hunza. *Pakistan. Radiation Protection Dosimetry, 153*(3), 390–399.

Alshahri, F., & El-Taher, A. (2019). Investigation of natural radioactivity levels and evaluation of radiation hazards in residential-area soil near a RasTanura refinery, Saudi Arbia. *Polish Journal of Environmental Studies, 28*(1), 25–34.

Bara, S. V., Arora, V., Chinnaesakki, S., Sartandel, S. J., Bajwa, B. S., Tripathi, R. M., & Puranik, V. D. (2012). Radiological assessment of natural and fallout radioactivity in the soil of Chamba and Dharamshala areas of Himachal Pradesh, India. *Journal of Radioanalytical and Nuclear Chemistry, 291*(3), 769–776.

Baykara, O., & Dogru, M. (2009). Determination of terrestrial gamma, ^{238}U, ^{232}Th and ^{40}K in soil along fracture zones. *Radiation Measurements, 44*(1), 116–121.

Butkus, D. (2006). *Jonizuojančioji spinduliuotė aplinkoje*. Vilnius: Technika *(in Lithuanian)*.

Butkus, D., & Konstantinova, M. (2008). Modelling vertical migration of ^{137}Cs in Lithuanian soils. *Journal of Environmental Engineering and Landscape Management, 16*(1), 23–29.

Butkus, D., Lebedytė, M., Lubytė, G., Matusevičius, K., & Mažeika, J. (2001). ^{137}Cs and ^{90}Sr in Soils of Lithuania. *Geochemistry International, 39*(7), 719–724.

Changizi, V., Shafiei, E., & Zareh, M. R. (2013). Measurement o ^{226}Ra, ^{232}Th, 1^{37}Cs and ^{40}K, and activities of wheat and corn products in Ilam Province–Iran and resultant annual ingestion radiation dose. *Iranian Journal of Public Health, 42*(8), 903–914.

Durusoy, A., & Yildirim, M. (2017). Determination of radioactivity concentrations in soil samples and dose assessment for

Rize Province, Turkey. *Journal of Radiation Research and Applied Science, 10*(4), 348–352.

Eke, C., Agar, O., & Boztosun, I. (2015). Dependence of activity concentration of natural and artificial radionuclides on depth in soil samples from Antalya in Turkey. *Journal of Physics: Conference Series, 590*(1), 012044.

Ibraheem, A. A., El-Taher, A., & Alruwaili, M. H. (2018). Assessment of natural radioactivity levels and radiation hazard indices for soil samples from Abha, Saudi Arabia. *Results in Physics, 11*, 325–330.

Lubytė, J., & Antanaitis, A. (2004). Migration of radionuclides in arable land of Lithuania. *Environmental Engineering and Landscape Management, 12*(1), 22–29.

Nedveckaitė, T. (2004). *Radiacinė sauga Lietuvoje*. Vilnius: Kriventa (*in Lithuanian*).

Nguelem, E. J. M., Ndontchueng, M. M., & Motapon, O. (2016). Determination of ^{226}Ra, ^{232}Th, ^{40}K, ^{235}U and ^{238}U activity concentration and public dose assessment in soil samples from bauxite core deposits in Western Cameroon. *Mekongtso Nguelemet al. SpringerPlus, 5*(1), 1253.

Ravisankar, R., Chandrasekaran, A., Vijayagopal, P., Venkatraman, B., Senthilkumar, G., Eswaran, P., & Rajalakshmi, A. (2012). Natural radioactivity in soil samples of Yelagiri Hills, Tamil Nadu, India and the associated radiation hazards. *Radiation Physics and Chemistry, 81*, 1789–1795.

Šidiškienė, D. (2001). Černobylio atominės elektrinės avarija. *Pasiruošimas branduolinei avarijai Lietuvoje*. Vilnius: Šveikatos aplinka, 2 priedas, pp. 1–7. (*in Lithuanian*).

Singh, S., Rani, A., & Mahajan, R. M. (2005). ^{226}Ra, ^{232}Th and ^{40}K analysis in soil samples from some areas of Punjab and Himachal Pradesh, India using gamma ray spectrometry. *Radiation Measurements, 39*(4), 431–439.

Song, G., Chen, D., Tang, Z., Zhang, Z., & Xie, W. (2012). Natural radioactivity levels in topsoil from the Pearl River Delta Zone, Guangdong, China. *Journal of Environmental Radioactivity, 103*, 48–53.

Tahir, S. N. A., Jamil, K., Zaidi, J. H., Arif, M., & Ahmed, N. (2006). Activity concentration of ^{137}Cs in soil samples from Punjab province (Pakistan) and estimation of gamma-ray dose rate for external exposure. *Radiation Protection Dosimetry, 118*(3), 345–351.

Tompson, A., Bruton, C., Pawloski, G., Smith, D., Bourcier, W., Shumaker, D., Kersting, A., Carle, S., & Maxwell, R. (2002). On the evaluation of groundwater contamination from underground nuclear tests. *Environmental Geology, 42*, 235–247.

Turhan, S., Köse, A., Varinlioğlu, A., Şahin, S. L., Arıkan, I., Oğuz, F., Yücel, B., & Özdemir, T. (2012). Distribution of terrestrial and anthropogenic radionuclides in Turkish surface soil samples. *Geoderma, 187–188*, 17–124.

United Nations Scientific Committee on the Effects of Atomic Radiation. **(2000)**. Report to the general assembly, with scientific annexes**.** United Nations Publications, https://www.unscear.org/docs/publications/2000/V1602702.pdf

United Nations Scientific Committee on the Effects of Atomic Radiation. (2008). Report to the general assembly, with scientific annexes. United Nations Publications, https://www.unscear.org/docs/publications/2008/UNSCEAR_2008_Report_Vol.II.pdf