ASSESSMENT OF NATURAL RADIONUCLIDES IN SOIL SAMPLES IN SPECIFIC AREAS ON THE TERRITORY OF LITHUANIA

M. PEČIULIENĖ, V. VASILIAUSKIENĖ, V. KLIMA, D. JASAITIS*

Vilnius Gediminas Technical University, Sauletekio al. 11, LT-10223, Vilnius, Lithuania
Corresponding author*: dainius.jasaitis@vgtu.lt

Received January 6, 2020

Abstract. The activity concentrations of soil samples collected from different locations of Lithuania were determined by using HPGe detector based on high-resolution gamma spectrometry system. The following three unique areas in Lithuania have been chosen for the research. Vilnius city is the capital of Lithuania and this is the city with the largest population in the country which is dominated by an anthropogenic environment. Juodkrantė is a recreation area located near the Baltic Sea and Curonian Spit which is included on the UNESCO World Heritage List and has a natural environment and minimal urbanization. Visaginas is special for the nuclear power plant (which is currently being decommissioned). It is the only nuclear power plant in the Baltic States built close to the border with Belarus on the shore of Drūkšiai Lake.

The activity concentrations of naturally occurring radionuclides ^{40}K, ^{226}Ra, and ^{232}Th have been measured in different brands of the soil samples taken from Vilnius and Juodkrantė districts and in the technogenic soil samples from the territory of Visaginas Nuclear Power Plant. Among all identified natural radionuclides, the highest level was that of ^{40}K, *i.e.* the mean value was 414 ± 71 Bq kg^{-1}, while the minimum level was that of ^{232}Th, *i.e.* the mean value was 4.8 ± 0.8 Bq kg^{-1}. The mean value of ^{226}Ra activity concentration was 13.9 ± 2.0 Bq kg^{-1}. Having measured the activity concentrations of naturally occurring radionuclides ^{232}Th, ^{226}Ra, and ^{40}K, the possible impact of natural radiation on a human has been assessed by calculating the absorbed dose rate (nGy h^{-1}). The average values of calculated absorbed dose rates in the samples were found 27.71 nGy h^{-1} in Vilnius, 23.25 nGy h^{-1} in Juodkrantė, and 28.65 nGy h^{-1} in Visaginas. In the paper, the annual effective dose (mSv) was also calculated: the minimum annual effective dose was found in Vilnius (0.034 mSv) and the maximum in Visaginas (0.035 mSv).

Key words: Ionizing radiation, natural radionuclides, nuclear power plant, activity concentration, absorbed dose rate, annual effective dose.

1. INTRODUCTION

The gamma radiation of radionuclides naturally occurring in the soils is one of the most important sources of external exposure. The important distinguishing feature of this source is its uneven distribution, *i.e.* its exposure differs a lot depending on the location. Due to this reason, it is important to carry out the most detailed research, control and identify the reasons causing higher exposure. The main factors causing the exposure of humans are the activity concentrations of natural radionuclides occurring in the soils and building materials as well as the time spent by a human outdoors and indoors exposed to this radiation.

The typical levels of activity concentrations of naturally occurring radionuclides depend on the types of the rocks that have formed the soils [1]. The higher levels of radionuclides are found in igneous rocks, while those of radionuclides found in sedimentary rocks are significantly lower. However, there are some exceptions: the activity concentrations of radionuclides found in schists and phosphates might be quite high. The researches aimed at determining the levels of activity concentrations of radionuclides naturally occurring in the soils and analysing the influence made by these activities on the rate of the dose absorbed in the air are carried out in many countries [2–5].

Accumulation of a major part of radionuclides in the soils is determined by their distribution in solid and liquid (brines) phase [6–8]. In order to be able to forecast the behaviour of the element in the soils, it is important to know its sorption abilities and the influence of the various environmental factors on the migration of a radionuclide.

Humans are exposed every day to the natural radionuclides which are ^{232}Th, ^{226}Ra, and ^{40}K. These are the main radioactive isotopes found in Earth rocks. The members of the radioactive decay chains of ^{232}Th (14 %), ^{235}U and ^{238}U (55.8 %), along with ^{40}K (13.8 %) are responsible for the main contributions to the dose from natural radiation [9, 10].

The highest levels of all naturally occurring radionuclides are those of ^{40}K. A radioactive potassium isotope ^{40}K is one of the main radionuclides which do not belong to radioactive families. Due to ^{40}K, a human is exposed to approximately 0.3 mSv annual radiation dose which is absorbed by a body together with non-radioactive isotopes of potassium (which is necessary for human vital activity). Potassium occurring in the earth has 0.011 % of radioactive isotope ^{40}K. This isotope is the main source of the natural exposure of a human body; each human has approximately 3.7×10^3 Bq of radioactive ^{40}K. In total, a human is internally and externally exposed to 180 μSv per year [11].

Due to different physical and geochemical properties of uranium- and thorium-series, radionuclides migrate in different velocities and different ways in natural environment [12]. The activity concentrations of ^{226}Ra may be lower than those of ^{238}U because the mobility of radium is greater than that of uranium. ^{226}Ra decay product is the gas element radon ^{222}Rn which emits from the soils, thus it has even more mobility. Therefore the quantities of ^{222}Rn decay products are lower than those of ^{226}Ra. Similar processes are also occurring among the members of ^{232}Th chain.

The radiation protection optimization principle requires the doses to which humans are exposed, and the number of exposed humans to be as low as reasonably achievable by taking into consideration social and economical conditions. The optimization principle also includes identification of the existing and potential radiation ways and sources. For many years the *United Nations Scientific Commission on Effects of Atomic Radiation* (UNSCEAR) has collected the data on various, *i.e.* both natural and artificial sources of exposure of a human from all over the world [13]. The aim is not only to determine the importance of these sources in different regions, countries and all over the world, but also to help in searching for the ways to reduce exposure of humans [14]. Identification of radiation sources, determination of the doses of their radiation, research on the regularities which cause the changes and distribution of these doses is a very important stage of radiation protection optimization. The variety of measures used to achieve these purposes is constantly increasing. One of the main reasons for this is the results obtained and regularities determined during the scientific research aimed at analysing radiation sources. The analysis of the sources of exposure of a human and regularities related to these sources is the relevant element of the research on the impact of ionizing radiation and search for the optimized measures of protection against the dangers arising from ionizing radiation [15, 16].

A human has a continuous impact on the environment; therefore it is relevant to assess the trends of ionizing radiation change. The anthropogenic assessment of natural ionizing radiation change is especially important in terms of environmental protection. The ionizing radiation changes were investigated in urbanized and non-urbanized territories as well as in the territory next to the former nuclear power plant. It has been chosen to perform the measurements in the geophysical locations which are mostly related to human activities and territories where anthropogenic activities are rather limited.

The aim of the paper is to carry out the research on a gamma ionizing radiation in urbanized and natural environments as well as sensitive territories (close to the former NPP), assess the changes in ionizing radiation caused by anthropogenic activity, determine the activity concentrations of naturally occurring radionuclides in the soils of different size distribution, and assess the contribution of natural radionuclides occurring in soils and building materials to the dose of the radiation emitted by them.

2. MEASUREMENT AND METHODS

2.1. THE LOCATION OF THE MEASUREMENT

The research was carried out in the territory of Lithuania. Lithuania is located in the central part of Europe, on the southeastern coast of the Baltic Sea. The country is divided into five regions and covers the area of 65.286 km^2. Lithuania shares borders with Latvia, Belarus, Poland and Russia (Kaliningrad Region). The following

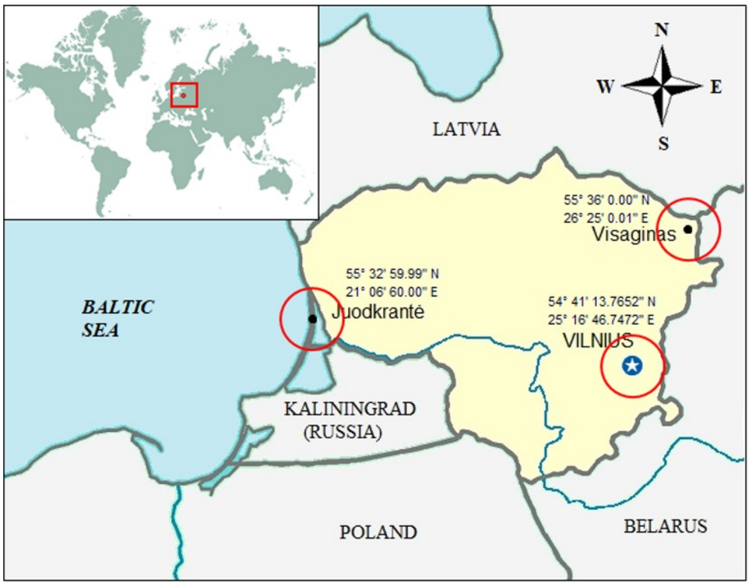

Fig. 1 – Sampling location.

three unique areas in Lithuania have been chosen for the research: Vilnius, Juodkrantė, and Visaginas (Fig. 1). Vilnius is the largest city of the country and the most urbanized area in Lithuania. This is the city with the largest population in the country and is dominated by an anthropogenic environment. Juodkrantė is characterized by a very clean environment; it is a recreation zone in the Curonian Spit which is included on the UNESCO World Heritage List. Juodkrantė has a natural environment and minimal urbanization. Visaginas is special because of the nuclear power plant (which is currently being decommissioned). It is the only nuclear power plant in the Baltic States built close to the border with Belarus on the shore of Lake Drūkšiai.

The western part of Lithuania is dominated by loam and sandy loam soils, the seashore by fine sand soil, the northern part by loam and clay, the eastern and southern part by sandy loam and sand, and the central part is dominated by sandy loam and loam.

Vilnius, as the whole southeastern part of Lithuania, is dominated by three types of soils, *i.e.* sand, sandy loam, and loam. The activity concentrations of naturally occurring radionuclides ^{232}Th, ^{226}Ra, and ^{40}K in soil have been measured by taking samples from different parts of Vilnius city.

Juodkrantė is the area which does not have a large cultural layer, active economic and industrial activities. A major part of the territory consists of forests and coastal sand and dunes. The condition of the beach depends on the geological structure of the underwater shore slope, the amount of sediment, and the lithodynamic situation. The part of the coastline above water consists of a sandy beach and shore dunes. The coast is dominated by fine sand. A part of the coast is covered with stones. The soil samples were taken from different areas of Juodkrantė town, *i.e.* from recreation zone (beach) and urbanized part of the town.

The samples in Visaginas were taken from the site which is located next to the former, closed Ignalina nuclear power plant. The construction of the third unit of former Ignalina NPP was started, but never finished in this territory, which later was demolished. The reinforced concrete structures were dismantled down to 1 m depth. The massive reinforced concrete structure foundations of 10 m depth were abandoned at the site. The whole territory is covered by the refilled technogenic soil except some places. The sediment-subsidized depth at the whole site varies from 0.2 m 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 third block and utility buildings of the former NPP were dismantled and laid in the trenches of the previous foundations. 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 deeper than 1 m was mixed with the constructional waste, namely concrete rubbish, reinforcement, plastics, aggregate, and timber. In some places below the above–mentioned soil, there is underlay sandy loam and fill with sand lenses and the layer from the Baltic period.

2.2. METHODOLOGY

The activity concentrations of naturally occurring radionuclides ^{232}Th, ^{226}Ra, and ^{40}K were determined experimentally in the samples of the dominant soil from Vilnius and Juodkrantė and in the samples of technogenic soil from the territory of Visaginas NPP. The samples were crushed, dried at 105 °C up to the constant weight and sealed in the cylindrical plastic measuring containers of 200 ml capacity (diameter 0.07 m, height 0.05 m) for four weeks to reach equilibrium ^{226}Ra and ^{232}Th and stabilization of their decay products. The gamma spectrometry system (Canberra Industries, USA) with the semiconductor HyperPure germanium (HpGe) detector has been used to determine the activity concentration of radionuclides.

HpGe detector have an energy resolution (FWHM) of 1.9 keV for 1.33 MeV gamma energy of ^{60}Co, relative efficiency – 30 %. Its sensor has high resolution and stability in time. Detector have high peak-to-Compton ratios of 58:1, which are expressed as the ratio of maximum counts in the peak channel to the average number of counts in the Compton plateau region. The detector surrounded by a 10 cm thick lead shield to reduce the background scattering, and cooled by liquid nitrogen to reduce leakage current detector caused by thermal noise. The integrated signal processor consists of a pulse height analysis system to transform pulses, which are finally collected by a computer-based multichannel analyzer (MCA), and analysis preformed with Genie 2000 software of Canberra.

For the calibration of the spectrometer ^{152}Eu + ^{137}Cs standards with the similar geometry as the soil samples were used. Calibration procedures and recommendations of the high and low resolution gamma-ray spectrometers are presented in [17].

The samples were measured for at least 90.000 s. Radionuclides were identified by the following energies: 186 keV of ^{226}Ra, 583 keV of ^{208}Tl, and 1460 keV of ^{40}K.

Radionuclide concentration A was calculated by the following formula (1):

$$A = \frac{S}{m \cdot t \cdot \eta_\gamma \cdot \varepsilon_E}, \quad (1)$$

where S is the spectral peak minus background (imp); ε_E is the efficiency of the gamma spectrometer; η_γ is the gamma quanta emission intensity; m is the mass of the sample (kg); t is the measurement time (s).

^{208}Tl is the decay product of ^{232}Th-series. Thus, ^{232}Th should be calculated (2):

$$A_{^{232}\mathrm{Th}} = 1.6 \times A_{^{208}\mathrm{Tl}}, \quad (2)$$

where $A_{^{232}\mathrm{Th}}$ is the ^{232}Th radioactive concentration of the sample (Bq kg^{-1}); $A_{^{208}\mathrm{Tl}}$ is the ^{208}Tl, radioactive concentration of the sample (Bq kg^{-1}).

3. RESULTS

Natural exposure depends on the quantities of natural radionuclides occurring in the earth's crust which differ due to the different composition of the soil. Although the radionuclide distribution in soil has been quite well studied, however the quantitative assessment of their activities is usually ambiguous. From a radiological point of view the soil studies are particularly important in anthropogenically affected locations. The intensity of natural radiation and its exposure depend on geological and geographical environment, therefore they differ in various regions. The studies have been carried out in the following locations of the Lithuanian territory: in the capital Vilnius, Curonian Spit and in the area next to the decommissioned nuclear power plant.

The activity concentrations of naturally occurring radionuclides ^{232}Th, ^{226}Ra, and ^{40}K in soil have been measured by taking samples from both highly urbanized areas and the areas less affected by the human activity as well as recreation zones. Table 1 represents the activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K in different regions of Lithuania.

The values of individual radionuclides varied within a wide range of values. Due to the different composition of soil and different quantities of depositions, the quantities of naturally occurring radionuclides in the samples vary up to 3 times.

Table 1

Activity concentration ($Bq\,kg^{-1}$) in different locations of Lithuania (M.V. $\pm\sigma$)

Location	Samples		^{232}Th	^{226}Ra	^{40}K
Vilnius	130	Range	0.3–18.4	2.1–35.1	263.2–710.1
		Mean	4.8 ± 1.2	12.9 ± 3.2	452.1 ± 112.4
Juodkrantė	27	Range	1.1–19.2	3.1–28.0	155.3–380.0
		Mean	5.0 ± 1.0	15.0 ± 2.0	319.0 ± 85.0
Visaginas	25	Range	0.7–21.3	4.3–37.4	185.2–620.7
		Mean	4.5 ± 0.8	13.7 ± 3.1	470.1 ± 93.0

^{232}Th varied from 0.3 to 21.3 $Bq\,kg^{-1}$, ^{226}Ra values varied from 2.1 to 37.4 $Bq\,kg^{-1}$, and ^{40}K values varied from 155.3 to 710.1 $Bq\,kg^{-1}$. The results are not distinguished by regions. The quite similar values of the researched radionuclides have also been found in the neighbouring countries of Lithuania [13, 18]. Table 2 represents the activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K in different locations of Baltic Sea region.

Table 2

Activity concentration ($Bq\,kg^{-1}$) in different locations of Baltic Sea region

Location	^{232}Th	^{226}Ra	^{40}K
Poland	4–77	5–120	110–970
Denmark	8–30	9–29	240–610
Estonia	5–59	6–31	140–1120
Russia	2–79	1–76	100–1400
Germany	7–134	5–200	40–1340

The detailed distributions of the activity concentrations of naturally occurring radionuclides ^{232}Th, ^{226}Ra, and ^{40}K in the researched different regions are provided in Fig. 2.

The activity concentration of ^{40}K in sand samples varied within a very wide range, *i.e.* from 120 to 566 $Bq\,kg^{-1}$. However, this is not the case with the sandy loam where the values varied only by approximately 7.5 % from the average value. Higher values of ^{40}K have been found in the samples of sandy loam and technogenic soil.

The activity concentrations of ^{226}Ra in soil vary from 2 to 37 $Bq\,kg^{-1}$. The activity concentration of this radionuclide is higher in the samples of sand and sandy loam.

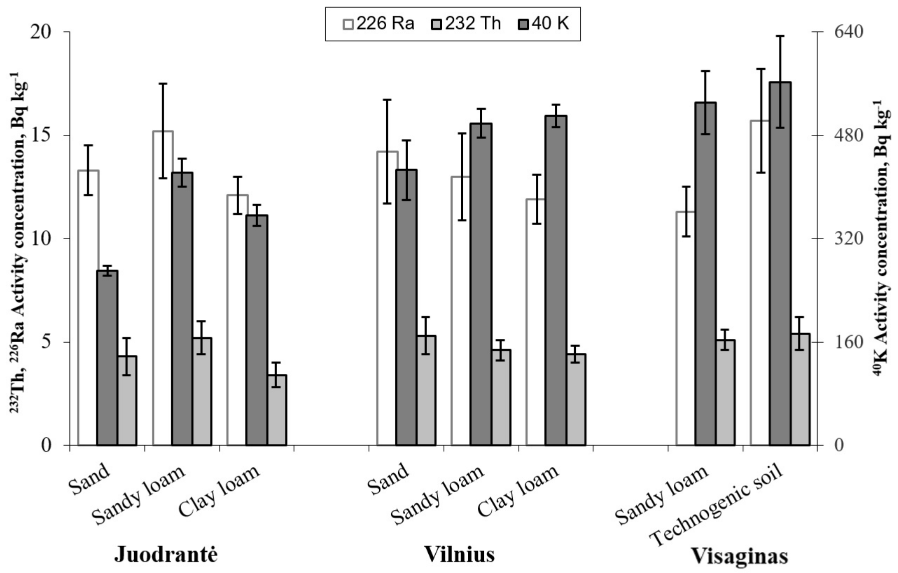

Fig. 2 – Distribution of activity concentrations of ^{40}K, ^{232}Th, ^{226}Ra in different regions of Lithuania.

The activity concentration of the decay product of ^{232}Th, *i.e.* ^{208}Tl, vary from 0.3 to 21 Bq kg^{-1}. The maximum values of ^{232}Th have been measured in sand and sandy loam samples, while the minimum values have been found in loam. It has been noticed that the higher activities of this radionuclides are found in the areas which are more affected by anthropogenic activity, while the lower values are measured in minimally urbanized areas such as parks, beaches and seashores.

From a radiological point of view, the studies of the technogenic soil in the territory of Visaginas decommissioned nuclear station are very relevant. In addition to natural soil, the technogenic soil contains high quantities of subsoil including large quantities of concrete, silicate and ceramic brick waste. It has been found in the studied soil samples that the values of activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are higher than the values of the same radionuclides found in the samples taken from other cities (Fig. 2).

Since the higher values of activity concentrations of radionuclides in technogenic soil are determined by the radioactivity of building materials in such soil, carrying out spectrometric assessment of the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in these materials, quite large quantities of them contributing to the additional exposure in the environment surrounding a human have been observed (Table 3).

The building materials have been studied from the radiological point of view. The materials which are mostly used in house building, *i.e.* silicate bricks, ceramic (clay) bricks, cement, concrete, finishes and timber, have been chosen for the study.

Table 3

Activity concentrations of building materials found in technogenic soil ($Bq\,kg^{-1}$)

Materials	Samples	Activity concentration (M.V. $\pm\sigma$)		
		^{232}Th	^{226}Ra	^{40}K
Brick (silicate)	15	14.9 ± 4.3	32.5 ± 8.1	749.3 ± 224.2
Brick (ceramic)	15	36.1 ± 4.3	68.3 ± 10.2	980.4 ± 208.1
Wood	18	2.3 ± 0.7	5.2 ± 1.6	38.4 ± 11.9
Concrete	18	34.7 ± 8.1	47.2 ± 9.2	543.4 ± 101.3
Gravel	10	3.7 ± 1.2	12.2 ± 3.7	296.1 ± 74.3
Gypsum	8	0.9 ± 0.4	1.9 ± 0.9	215.4 ± 25.2
Cement	11	4.0 ± 1.2	41.5 ± 12.4	157.3 ± 47.1

The results of the study of the activity concentration of ^{226}Ra in building material show that the values of the activity concentration of this radionuclide vary quite widely. It has been established that the lowest quantities of ^{226}Ra are found in plaster, while sand contains up to 10 times higher quantities of this radionuclide. The activity concentration of ^{232}Th in the studied building materials has the lowest value among all identified radionuclides. The results of the studies show that the lowest values of the activity concentration of this radionuclide have been found in timber, gravel and cement samples. The building materials contain the highest quantities of durable radionuclide ^{40}K. The radiation of this particular radionuclide has the biggest impact on external exposure. The values of the activity concentration of this radionuclide vary in a very large range. The radiation induced by the radionuclides contained in an artificial environment is approximately 1.3 times higher than that induced by the radionuclides in soil. This fact reflects the influence of anthropogenic activity on a natural radiation.

Having carried out the spectrometric analysis of the environment surrounding a human, the contribution of the different radionuclide contained in soil to the exposure has been assessed. In order to assess the possible impact of natural radiation on a human, by taking into consideration the results of the studies carried out in different locations, the absorbed dose rate ($nGy\,h^{-1}$) has been calculated by using the Monte Carlo method [15].

Conversion factors to transform activity concentrations A_K, A_{Ra}, A_{Th} of K, Ra, Th, respectively, in absorbed dose rate (D) at 1 m above the ground (in $nGy\,h^{-1}$ by $Bq\,kg^{-1}$) was calculated by Monte Carlo method as [13]:

$$D = 0.0417 A_K + 0.462 A_{Ra} + 0.604 A_{Th}, \tag{3}$$

where A_K, A_{Ra}, A_{Th} are the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in

$\mathrm{Bq\,kg^{-1}}$.

From Table 1, the average values of calculated absorbed dose rate in samples were found 27.71 $\mathrm{nGy\,h^{-1}}$ in Vilnius, 23.25 $\mathrm{nGy\,h^{-1}}$ in Juodkrantė, and 28.65 $\mathrm{nGy\,h^{-1}}$ in Visaginas (Table 4).

Table 4

Absorbed and annual efective dose rates in different locations of Lithuania

Location	Absorbed dose rate ($\mathrm{nGy\,h^{-1}}$)	Annual efective dose (mSv)
Vilnius	27.71	0.034
Juodkrantė	23.25	0.029
Visaginas	28.65	0.035

The indoor and outdoor occupancy factor and the conversion coefficient from absorbed dose in air to effective dose must be considered for estimation of the annual effective doses. Equivalent of the annual estimated average effective dose which a member was exposed to was determined by using a conversion factor of 0.7 $\mathrm{Sv\,Gy\,h^{-1}}$. This factor is used to convert the absorbed rate to the annual effective dose with an outdoor occupancy of 20 % and 80 % for indoor environment [19, 20]. The outdoor annual effective doses are established as follows [21]:

$$\mathrm{AED} = D \times t \times 0.2 \times F, \tag{4}$$

where AED – Annual effective dose (mSv); D – absorbed dose rate ($\mathrm{nGy\,h^{-1}}$), t – is the duration of the exposure (8760 h), F – conversion factor of 0.7 $\mathrm{Sv\,Gy^{-1}}$.

The annual effective dose from Vilnius, Juodkrantė, and Visaginas samples were calculated. The minimum annual effective dose were found in Vilnius 0.034 mSv and the maximum in Visaginas 0.035 mSv. In Juodkrantė the annual effective dose was found 0.029 mSv. The results show that the maximum annual effective dose was found in technogenic material (Visaginas).

The annual effective dose from Visaginas technogenic soil are higher than other samples in Vilnius and Juodkrantė. The lowest dose was found in Vilnius, however, all these samples satisfy the safety criteria for radiation safety point of view and hence these soil and technogenic soil samples do not pose any environment and health hazard problems. The calculated annual effective dose is below the limit of 1.0 $\mathrm{mSvy^{-1}}$ recommended by the International Commission on Radiological Protection, for the general public.

4. CONCLUSIONS

In order to evaluate the impact of natural ionizing radiation on a human in an urbanized and natural environment, it is necessary to control the activity concentrations of naturally occurring radionuclides and their contribution to additional exposure. Carrying out comprehensive radiological studies of soil, it has been established that the soil contains the highest quantities of ^{40}K, *i.e.* mean value 414 ± 71 Bq kg^{-1}, and the lowest quantities of ^{232}Th, *i.e.* mean value 4.8 ± 0.8 Bq kg^{-1} from all identified natural radionuclides.

^{226}Ra activity concentration mean value is 13.9 ± 2.0 Bq kg^{-1}. The higher values of ^{232}Th, ^{226}Ra, and ^{40}K have been found in sand and sandy loam samples. It has been determined that the higher concentrations of radionuclides are found in the technogenic soil within a sensitive area (next to decommissioned nuclear power station) resulting from radioactivity of the building materials in soil. Having assessed the possible additional exposure induced by the radioactivity of the building rubble materials contained in soil, it has been established that the absorbed dose rate and annual effective dose increase up to 1.2 times. However, they do not exceed the maximum permissible rates of natural exposure. The studied areas may be considered as safe from a radiological point of view.

REFERENCES

1. M. Shirav (Schwartz), *Some remarks on high natural background radiation terrains, geology and public health in Israel and adjacent countries*, IRPA 11 congress papers, 1–5 (2004).
2. J. Al-Jundi, W. Salah, M. S. Bawaaneh, and F. Afaneh, *Exposure to radiation from the natural radioactivity in Jordanian building materials*, Radiat. Prot. Dosim. **118**(1), 93–96 (2006).
3. F. B. Masok, P. L. Masiteng, and I. D. Jwanbot, *Natural radioactivity concentrations and effective dose rate from Jos tin mining dumpsites in Ray-field*, Nigeria, J. Environ. Earth Sc. **5**(12), 51–55 (2015).
4. D. A. E. Darwish, K. T. M. Abul-Nasr, and A. M. El-Khayatt, *The assessment of natural radioactivity and its associated radiological hazards and dose parameters in granite samples from South Sinai, Egypt*, J. Radiat. Res. Appl. Sc. **8**(1), 17–25 (2015).
5. F. B. Masok, P. L. Masiteng, R. D. Mavunda, P. P. Maleka, and H. Winkler, *Measurement of radioactivity concentration in soil samples around phosphate rock storage facility in Richards Bay, South Africa*, J. Radiat. Res. Appl. Sc. **11**(1), 29–36 (2018).
6. P. Blanco Rodríguez, F. Vera Tom, and J. C. Lozano, *Assessment of the vertical distribution of natural radionuclides in a mineralized uranium area in south-west Spain*, Chemosphere **95**, 527–534 (2014).
7. C. Dumant, S. Stauton, *Reduced adsorbtion of caesium on clay minerals caused by various humic substances*, J. Environ. Radioact. **46**(2), 187–200 (1999).
8. L. Monte, R. Perianez, P. Boyer, J. T. Smith, and J. E. Brittain, *The role of physical processes controlling the behaviour of radionuclide contaminants in the aquatic environment: a review of state-of-the-art modelling approaches*, J. Environ. Radioact. **100**(9), 779–784 (2009).
9. P. Ele Abiama, P. Owono Ateba, G. H. Ben-Bolie, F. H. P. Ekobena, and T. El Khoukhi, *High background radiation investigated by gamma spectrometry of the soil in the southwestern region of Cameroon*, J. Environ. Radioact. **101**(9), 739–743 (2010).

10. E. O. Joshua, J. A. Ademola, M. A. Akpanowo, O. A. Oyebanjo, and D. O. Olorode, *Natural radionuclides and hazards of rock samples collected from Southeastern Nigeria*, Radiat. Meas. **44**(4), 401–404 (2009).
11. IAEA. International Atomic Energy Agency, *Extent of environmental contamination by naturally occurring radioactive material (NORM) and technological options for mitigation*, IAEA Technical Reports Series No 419, Vienna, 1–208 (2003).
12. S. B. Chena, Y. G. Zhua, and Q. H. Huc, *Soil to plant transfer of ^{238}U, ^{226}Ra and ^{232}Th on a uranium mining-impacted soil from southeastern China*, J. Environ. Radioact. **82**(2), 223–236 (2005).
13. UNSCEAR. United Nations Scientific Committee on the Effects of Atomic Radiation, *Sources and effects of ionizing radiation*. UNSCEAR 2000 report to the General Assembly, Vol 1. United Nations, New York, 1–654 (2000).
14. EC, *Council directive 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation*, Off. J. Eur. Union, L **13**(1), 1–73 (2013).
15. A. A. Ibraheem, A. El-Taher, and M. H. Alruwaili, *Assessment of natural radioactivity levels and radiation hazard indices for soil samples from Abha, Saudi Arabia*, Results Phys. **11**, 325–330 (2018).
16. H. Florou, G. Trabidou, and G. Nicolaou, *An Assessment of the External Radiological Impact in Areas of Greece with Elevated Natural Radioactivity*, J. Environ. Radioact. **93**(2), 74–83 (2007).
17. A. Luca, B. Neacsu, A. Antohe, and M. Sahagia, *Calibration of the high and low resolution gamma–ray spectrometers*, Rom. Rep. Phys. **64**(4), 968–976 (2012).
18. K. M. Thabayneh, M. M. Jazzar, *Natural Radioactivity Levels and Estimation of Radiation Exposure in Environmental Soil Samples from Tulkarem Province-Palestine*, Open J. Soil Sci. **2**(1), 7–16 (2012).
19. G. Alzubaidi, F. B. Hamid, and I. Abdul Rahman, *Assessment of natural radioactivity levels and radiation hazards in agricultural and virgin soil in the state of Kedah, North of Malaysia*, Sci. World J. **1**, 1–9 (2016).
20. R.G. Sonkawade, K. Kant, S. Muralithar, R. Kumar, and R.C. Ramola, *Natural radioactivity in common building construction and radiation shielding materials*, Atmos. Environ. **42**(9), 2254–2259 (2008).
21. B. M. Moharram, M. N. Suliman, N. F. Zahran, S. E. S. Shennawy, and A. R. El Sayed, *External exposure doses due to gamma emitting natural radionuclides in some Egyptian building materials*, Appl. Radiat. Isot. **70**(1), 241–8 (2012).