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Natural radionuclides in the soil of the Vilnius city and assessment of their hazard

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The radioisotopic composition of different soil types of the Vilnius city (54°40'N, 25°20'E), the capital and the biggest city of Lithuania, was identified. The specific activities of natural radionuclides in the soil were measured with a gamma spectrometric system containing a HPGe detector. The specific activities of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in samples of different granulometric composition were measured. The maximum values of ^{40}K were identified in the southeastern and eastern parts of the city (about 479 Bq/kg) and the minimum values in the northern and western parts (about 200 Bq/kg). The maximum values of ^{226}Ra and ^{232}Th specific activities were measured in the southern and southeastern regions (23 Bq/kg and 8 Bq/kg, respectively) and the minimum values in the central part of the city (7 Bq/kg and 3 Bq/kg, respectively). The contribution to the external equivalent dose rate of separate gamma emitters ^{226}Ra, ^{232}Th and ^{40}K was estimated. In the Vilnius city, the radiation of the equivalent dose rate makes up on average 12 nSv/h due to ^{40}K present in the soil, 6 nSv/h due to ^{226}Ra, 3 nSv/h due to ^{232}Th.

Key words: sand, sandy loam, clay loam, natural radionuclides, equivalent dose rate, specific activity, Lithuania

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INTRODUCTION

Recently, the attention paid to artificial radionuclides has been very conspicuous, whereas attention to radionuclides of natural origin has been less, though it is known that the contribution of artificial radionuclides in our environment is much smaller. Natural radionuclides in soil generate a significant component of the background radiation exposure of the population (Miah et al., 1998).

A significant part of the dose in the form of natural sources comes from terrestrial gamma radionuclides (UNSCEAR, 2000). The most widely spread radionuclides of natural origin come from the uranium (^{238}U), thorium (^{232}Th) and actinium (^{235}Ac) families as well as from potassium (^{40}K). The exposure of the population from natural sources depends on the natural radionuclides found in the underground, their quantities depending on soil composition. The distribution of radionuclides in soil has been very widely explored, yet the quantative evaluation of their activities is frequently ambiguous, and this is because of soil being a complicated component of nature. The behavior of many radionuclides in soil is caused by the processes of their distribution between two main phases – solid and liquid (leaches of soil). These processes are formed by reversible or irreversible absorption-dissipation, precipitation-dissolution, coagulation-peptide formation processes of radionuclides. Taking into consideration the complexity of all these processes, the quantitative estimations of the main natural gamma emitters found in the soil of the Vilnius city were conducted and their contribution to irradiation were estimated. Such researches were carried out also in other regions of Lithuania (Vilkaviškis, Prienai, Marijampolė, Lazdijai, Varėna and Ignalina) (Mažeika, 2002).

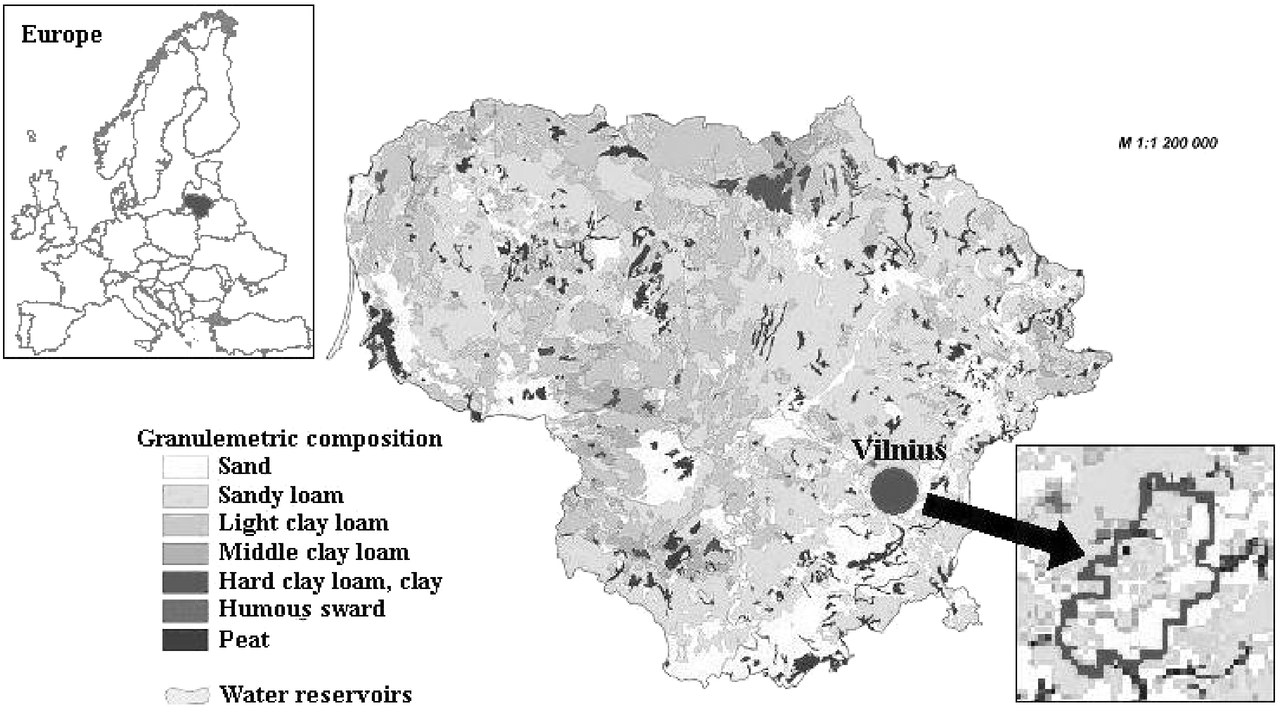

Fig. 1. The map of granulometric composition of Lithuania's soil
1 pav. Lietuvos dirvožemio granuliometrinės sudėties žemėlapis

The Republic of Lithuania lies on the eastern coast of the Baltic Sea. In the north the country borders on Latvia, in the east on Belarus, and in the south on Poland and Kaliningrad Region of the Russian Federation. Vilnius is the capital of Lithuania and the biggest city in the country (about 550 thousand people). According to the composition of the soil, clay loam and sandy loam dominate in the west of Lithuania, fine sand prevailing in the seaside, clay loam and clay in the north, sandy loam and sand in the eastern and southern parts, and sandy loam and clay loam in the central part of Lithuania (Fig. 1) (Soviet Lithuania map of the soil, 1985).

The intensity of natural radiation and the resulting exposure depend on the geological and geographical environment, therefore in various regions it is different (UNSCEAR, 2000).

The aims of the current research were to estimate the radioisotopic composition in different types of soil in the Vilnius city; to measure the specific activities of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K found in samples of different granulometric composition; to evaluate the contribution of separate gamma emitters (^{226}Ra, ^{232}Th and ^{40}K) to their external equivalent dose rate.

The study was carried out in the Vilnius city where, like in all southeastern Lithuania, the prevailing types of soil are sandy loam and sand. Samples of different types of soil were taken for this research, and specific activities of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K were measured and their external equivalent dose rate was estimated.

METHODS

The samples were taken in the southern, western, northern and central parts of the Vilnius city and divided into three groups according to the type of soil – sand, sandy loam, clay loam; 72 samples from the surface layer of the soil (0–5 cm) were taken.

A metal ring 150 mm in diameter and 50 mm high was used to take soil samples; it was hammered into the ground and the ground from the bottom was lifted up with a spade. The samples were transported in plastic bags to the laboratory. Then they were weighed and placed into 200 ml vessels in the laboratory. Dry soil was granulated, sieved out, poured into a measurement vessel and weighed.

To identify the specific activity of natural radionuclides in the soil, the activity of the samples was measured with a gamma spectrometric system (CANBERRA) equipped with a semiconductor HPGe detector (resolution 2 keV, efficiency 15%). The detector's sensor was of a high differential power as it was constantly freezed with liquid nitrogen. The spectrometer was calibrated for a 200 ml vessel. Solid ^{152}Eu +^{137}Cs standards of definite activity of different densities were used for calibration.

The specific activity of the main gamma emitters in the soil was identified using exposures of 24–72 hour sample measurements. Radionuclides were identified according to the following energy lines: ^{226}Ra – 186 keV, ^{208}Tl – 583 keV, ^{137}Cs – 662 keV, ^{40}K – 1460 keV. ^{208}Tl is the product of decay of ^{232}Th, therefore their ratio of specific activity was taken equal to 1.6 and was respectively calculated (Aycik, Erkan, 1997).

The specific activity of the radionuclides was recalculated according to the formula:

$$A = \left(\frac{S}{t} - \frac{S_f}{t_f} \right) \Big/ m \cdot \varepsilon \cdot \eta ,$$

where S is the area of the peak of the measured material of the radionuclide, imp/s; S_f is the area of the peak of background radiation, imp/s; t is the measurement time of the sample, s; ε is the product of the energetic and geometric efficiency of the semiconductor spectrometer; η is the quantum output of radionuclide decay; m is the mass of the measured sample, kg.

In the course of measurements, the background of gamma radiation was periodically measured. Its level in the course of measurements was more or less steady. The exposures of the measured samples were selected so that changes of the background would not make up more than 4% of the size of their signal.

RESULTS

Three types of soil – sand, sandy loam and clay loam – prevail in the Vilnius city. The specific activities of natural radionuclides ^{232}Th, ^{226}Ra and ^{40}K found in the soil, taking samples from different parts of Vilnius, were estimated. The statistical treatment of the specific activities of natural radionuclides in the soil is presented in Table 1.

^{40}K was found to show the highest specific activity out of all the radionuclides studied; its contribution to the external equivalent dose rate was the largest. High correlation coefficients between the equivalent dose rate in the air layer close to the ground surface with the main ^{40}K gamma emitters and ^{226}Ra specific activities in the soil (0.66 and 0.70, respectively) were identified. Meanwhile, a low negative correlation (-0.24) was noted between the equivalent dose rate and the specific activity of ^{232}Th in the soil. Also, in 1992–1995 the specific activity in the soil of the main gamma emitters, ^{40}K and ^{226}Ra, and the equivalent dose rate close to the ground surface above the location of the sample were measured. The results showed that the dose rate in the air layer close to the ground surface was mostly influenced by ^{40}K found in the soil (Lietuvos…, 1999). The external equivalent dose rate, influenced by radiation of the radionuclides present in the soil, comprises 35–64% of the total equivalent dose rate in the air layer close to the ground surface, in which ^{40}K takes a significant part (on average 33 %); ^{226}Ra and its decay products make up on average 12%, ^{232}Th and its decay products making 6% (Konstantinova, Butkus, 2003).

Due to different soil composition and uneven amount of deposits, the content of natural radionuclides in the samples varied up to three times. Fig. 2 illustrates the distribution of natural radionuclides ^{232}Th, ^{226}Ra and ^{40}K in the soil samples.

The highest content of natural radionuclides was found in sand, but it depended also on the location the sample was taken from. We could not strictly distinguish the type of soil which, depending on the content of natural radionuclides in it, was more or less active.

In the samples of sand the values of the specific activity of ^{40}K varied in a wide range (from 479 to 200 Bq/kg), whereas in clay loam these values varied within 7.5% from the average. Higher values of ^{40}K were identified in the southeastern and eastern and smaller in the northern and western parts of the city.

The values of the specific activity of ^{226}Ra in the soil of the Vilnius city varied similarly to ^{40}K, i. e. from 7 to 23 Bq/kg. The specific activity of this radionuclide was higher in the southern and southeastern regions, the values being smallest in the central part of the city.

The values of the specific activity of ^{208}Tl, a decay product of ^{232}Th, ranged from 3 to 8 Bq/kg. They

Table 1. **Specific activities of natural radionuclides in the soil of Vilnius**
1 lentelė. **Gamtinės kilmės radionuklidų savitieji aktyvumai Vilniaus miesto dirvožemyje**

Radionuclide	Specific activity, Bq/kg				
	Maximum value	Minimum value	Average value	Median	Mean square deviation
^{232}Th	8.5	3.2	4.8	5.0	± 1.2
^{226}Ra	23.3	7.0	12.9	12.5	± 3.2
^{40}K	479.3	146.1	263.1	245.5	± 67.4

Table 2. **Specific activities of natural radionuclides in different types of soil**
2 lentelė. **Gamtinės kilmės radionuklidų savitieji aktyvumai skirtingų tipų dirvožemyje**

Soil type	Number of the samples	Specific activity, Bq/kg					
		^{40}K		^{226}Ra		^{232}Th	
		Average	Standard deviation	Average	Standard deviation	Average	Standard deviation
Sand	24	284.0	±46.3	14.2	±2.5	5.3	±0.9
Sandy loam	21	272.1	±22.2	13.0	±2.1	4.6	±0.5
Clay loam	27	232.0	±17.3	11.9	±1.2	4.4	±0.4

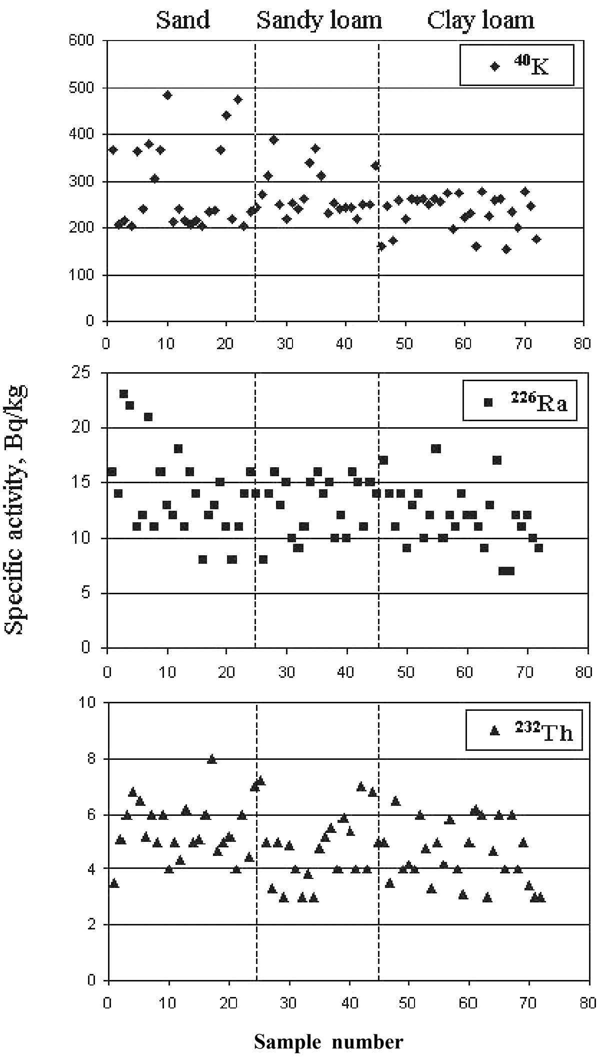

Fig. 2. Specific activities of ^{232}Th, ^{226}Ra and ^{40}K in soil samples of different granulometric composition

2 pav. ^{40}K, ^{226}Ra ir ^{232}Th savitieji aktyvumai skirtingos granuliometrinės sudėties dirvožemio bandiniuose

increased in the southern and southeastern parts, and the smallest values were identified in the central part of the city.

Table 2 presents a statistic treatment of the specific activities of the main gamma emitters in different types of soil.

The external equivalent dose rate caused by natural radionuclides in soil was estimated, because it directly depends on the specific activity of radionuclides in soil.

The equivalent dose rate close to the ground surface, caused by the ionizing radiation of radionuclides in soil, was identified according to the specific activity of the main natural gamma emitters – the specific activity of ^{232}Th, ^{226}Ra and ^{40}K in the cultivable and uncultivable land of Vilnius.

While calculating the external equivalent dose rate of natural radionuclides and considering that they are distributed equally in the soil layer of unlimited thickness, the dose rate was calculated:

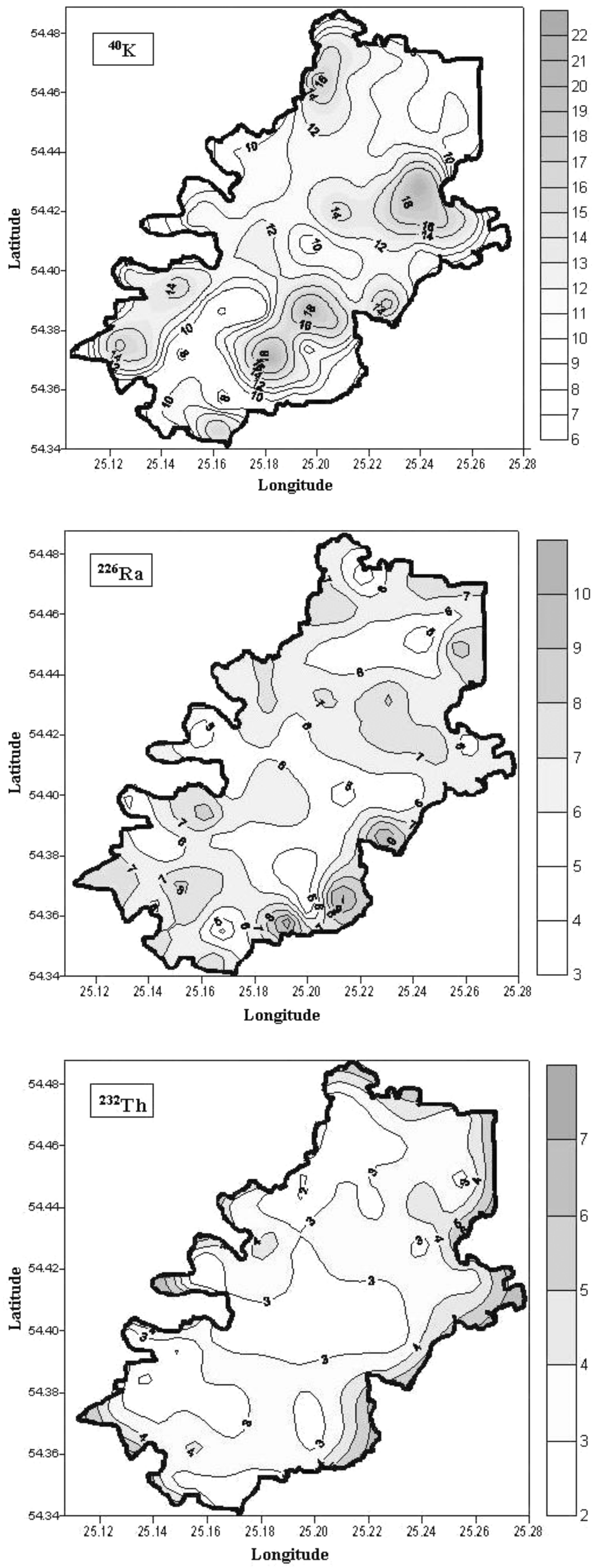

Fig. 3. Distribution of the external equivalent dose rate caused by radiation of ^{40}K, ^{226}Ra ir ^{232}Th found in the soil in the territory of Vilnius, nSv/h

3 pav. Išorinės lygiavertės dozės galios, sukeltos dirvožemyje esančių ^{40}K, ^{226}Ra ir ^{232}Th spinduliuočių, pasiskirstymas Vilniaus miesto teritorijoje (nSv/h)

$$\dot{H} = \left(\frac{2\pi \cdot K_\gamma \cdot A}{\mu_{ms}} \right) \cdot E \cdot \mu_m \cdot d ,$$

where $\dot{H}$ is the external equivalent dose rate; $K\gamma$ is the gamma radiation constant, Sv·m^2·s^{-1}·Bq^{-1}; A is the specific activity of a radionuclide in the surface layer of soil, Bq-kg^{-1}; μ_m and μ_{ms} are mass energy transfer coefficients of gamma radiation in the air and in the soil, respectively, m^2·kg^{-1}; d is the thickness of the air layer between the ground surface and the measurement point, multiplied by air density, kg·m^{-2}; E is the integral function of gamma radiation in the air and in the soil, which is expressed by the standard function E_n and depends on the mass energy transfer coefficients of air and soil.

The mass energy transfer coefficient of radiation μ_{ms} (Lebedytė, Morkūnas, Butkus, 1999), used for calculations, for dry soil was identified according to the type of soil.

The distribution isolines of the external equivalent dose rate caused by radiation of the natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th are presented in Fig. 3.

In the Vilnius city, the radiation of the equivalent dose rate makes up 12 nSv/h due to ^{40}K present in the soil, 6 nSv/h are due to ^{226}Ra, 3 nSv/h due to ^{232}Th. The largest external equivalent dose rate caused by the ionizing radiation of ^{40}K was identified in the southeastern and eastern parts of Vilnius (21 nSv/h). The smallest values of external equivalent dose rate caused by radiation of ^{40}K were identified in the northern and western parts of Vilnius (6–8 nSv/h). The soil type and its radioisotopic composition can predetermine such quite a big distribution of values.

The smallest external equivalent dose rate caused by the ionizing radiation of ^{226}Ra and ^{232}Th present in soil, was identified in the central part of the city (5 nSv/h and 3 nSv/h, respectively). The prevailing type of soil in this part of the city is sandy loam, which contains lower levels of natural radionuclides in its composition. The largest contribution of ^{226}Ra to its external equivalent dose rate was identified in the southern and southeastern regions of Vilnius (roughly 9–10 nSv/h). The prevailing soil type in this part of the city is sand.

CONCLUSIONS

The specific activity of the main natural gamma emitters ^{40}K, ^{226}Ra and ^{232}Th in different types of soil in the Vilnius city was measured. The values of the specific activity of all these radionuclides in the soil are distributed unevenly. The highest values of ^{40}K were identified in the southeastern and eastern parts of the city (sand and sandy loam prevails) and the smallest in the northern and western parts (sandy loam and clay loam prevail). The specific activity of ^{226}Ra is higher in the southern and southeastern regions (sand and sandy loam prevail), the smallest values being in the central part of the city (clay loam prevails). The highest values of the specific activity of ^{232}Th were measured in the southern and southeastern parts (sand and sandy loam prevail) and the smallest in the central part (clay loam prevails).

The contribution to the external equivalent dose rate of separate gamma emitters ^{40}K, ^{226}Ra and ^{232}Th was evaluated. In the Vilnius city, the equivalent dose rate of ^{40}K radiation makes up 12 nSv/h, radiation of ^{226}Ra making 6 nSv/h and of ^{232}Th 3 nSv/h in the average equivalent dose rate in the air layer close to the ground surface.

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GAMTINIAI RADIONUKLIDAI VILNIAUS MIESTO DIRVOŽEMYJE IR JŲ NULEMTOS APŠVITOS ĮVERTINIMAS

S a n t r a u k a

Darbe ištirta Vilniaus miesto (54°40'N, 25°20'E), Lietuvos sostinės ir didžiausio miesto Lietuvoje, skirtingų tipų dirvožemio radioizotopinė sudėtis. Gamtinės kilmės radionuklidų savitieji aktyvumai nustatyti naudojantis gama spektrometrine sistema su HPGe puslaidininkiniu detektoriumi. Išmatuoti gamtinės kilmės ^{226}Ra, ^{232}Th ir ^{40}K radionuklidų savitieji aktyvumai skirtingos granuliometrinės sudėties bandiniuose. Didžiausios ^{40}K vertės nustatytos pietrytinėje ir rytinėje miesto dalyse (apie 479 Bq/kg), mažiausios – šiaurinėje ir vakarinėje dalyse (apie 200 Bq/kg). ^{226}Ra bei ^{232}Th savitojo aktyvumo didžiausios vertės išmatuotos pietinėje ir pietrytinėje Vilniaus dalyse (atitinkamai 23 Bq/kg ir 8 Bq/kg), mažiausios – centrinėje miesto dalyje (atitinkamai 7 Bq/kg ir 3 Bq/kg). Apskaičiuotas atskirų gama spinduliuočių ^{226}Ra, ^{232}Th ir ^{40}K indėlis jų nulemtai išorinei lygiavertės dozės galiai. Vilniaus mieste dėl dirvožemyje esančio ^{40}K spinduliuotės lygiavertės dozės galia vidutiniškai sudaro 12 nSv/h, dėl ^{226}Ra – 6 nSv/h, o dėl ^{232}Th – 3 nSv/h.

Милда Пячулене, Дайнюс Ясайтис, Алоизас Гиргждис

ПРИРОДНЫЕ РАДИОНУКЛИДЫ В ПОЧВАХ Г. ВИЛЬНЮСА И ОЦЕНКА ИХ ОБЛУЧАЮЩЕЙ СПОСОБНОСТИ

Резюме

В работе определен радиоизотопный состав различных типов почв г. Вильнюса (54°40'N, 25°20'E) – столицы и крупнейшего города Литвы. Удельная активность природных радионуклидов определена с помощью гамма-спектрометрической системы с полупроводниковым детектором HPGe. Измерена удельная активность природных радионуклидов ^{226}Ra, ^{232}Th и ^{40}K в образцах различного гранулометрического состава. Наивысшая концентрация ^{40}K получена в юго-восточной и восточной частях города (около 479 Bq/kg), а наинизшая – в северной и западной частях (около 200 Bq/kg). Наибольшие значения удельной активности ^{226}Ra и ^{232}Th измерены в южной и юго-восточной частях города (23 и 8 Bq/kg соответственно), наименьшие – в центральной части города (7 и 3 Bq/kg соответственно). Рассчитана суммарная мощность гамма-излучения ^{226}Ra, ^{232}Th и ^{40}K, а также каждого радионуклида в отдельности. В Вильнюсе мощность облучения ^{40}K, находящегося в почве, составляет 12 nSv/h, ^{226}Ra – 6 nSv/h, а ^{232}Th – 3 nSv/h.