

SPECIFIC ACTIVITY OF RADIONUCLIDES IN SCOTS PINE (*PINUS SYLVESTRIS* L.) WOOD ON STABATIŠKĖS SITE OF IGNALINA NPP

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Abstract. A near surface repository for low and intermediate-level short-lived radioactive waste is being constructed on Stabatiškės site which is located in the territory of Visaginas Municipality near to Ignalina Nuclear Power Plant. If the specific activity of radionuclides in environmental objects were known before the construction of the repository, it would be possible to measure potential irradiance caused by the repository during its operation.

This article researches the specific activities of natural (²²⁶Ra, ²³²Th, ⁴⁰K) and artificial (¹³⁷Cs) radionuclides in Scots pine (*Pinus sylvestris* L.) wood on the Stabatiškės site before the construction of the repository. Having measured the specific activities in the ashes of annual tree rings, the concentration and distribution of radionuclides in Scots pine wood was assessed.

It was determined that there is no linear dependence between the change of pine biomass and the age of the tree. The change of biomass might have been caused by favourable and unfavourable climatic conditions and by atmospheric pollution.

The article presents curves showing the interrelation of the change of pine biomass and specific activities of natural (²²⁶Ra, ²³²Th, ⁴⁰K) as well as artificial (¹³⁷Cs) radionuclides.

Keywords: natural and artificial radionuclides, annual rings, the specific activity of radionuclides, Scots pine (*Pinus sylvestris* L.), correlation coefficients, the change of biomass, Stabatiškės site

1. Introduction

Two types of radionuclides, namely natural and artificial, exist in the environment. Natural radionuclides are found in rocks and soil and also they are a cosmic radiation. The most widely known natural radionuclides are members of uranium (²³⁸U), thorium (²³²Th) actinide series and potassium (⁴⁰K). The average concentration of radium (²²⁶Ra) in soil is 3–4 Bq·kg⁻¹, ²³⁸U – about 26 Bq·kg⁻¹, and ⁴⁰K – less than 600 Bq·kg⁻¹ (Beinaravičius 2005).

Artificial radionuclides are a product of human activities. The main sources of artificial radionuclides in the environment are testing of nuclear weapons and the nuclear fuel cycle, especially its accidents (Yvanov *et al.* 1997). After the technogenic nuclear accident in Chernobyl NPP, the total emission of the radioactive content was about 90 mCi (Pliopaitė Bataitienė 2006).

In the environment the migration of radionuclides is taking place together with the dusty air stream. Radionuclides settle on the ground and on the leaves of plants (Щеглов и Цветнова 2001).

The primary absorbers of radioactive pollution in the environment are plants. Radionuclides get into a tree

from the atmosphere and soil through its root system (Цыбулька *et al.* 2004).

Radionuclides get into the soil from the atmosphere and they are washed from the branches and leaves of trees as well. In the soil, due to diffusion and convective transfer, radionuclides migrate, reach roots and through them pass into plants (Butkus and Pliopaitė Bataitienė 2006; Kagawa *et al.* 2002).

The absorption through the organs of photosynthesis makes only about 10 % of the total amount of absorbed radionuclides. Researches show that the absorption of radionuclides through roots may be even up to 200 times higher than the absorption through leaves (Knapickas and Raupelienė 2004; Buzinny *et al.* 2000).

Radionuclides have different biologic, toxic and genotoxic impact on plants; they can disorder the physiological processes in plants, stop cell division, and reduce the vitality and reproduction of plants (Evseeva and Geraskin 2000; Marčiulionienė *et al.* 2007). Moreover, land plants are a very important source of polluted elements which get into a human body from food.

Trees protect natural environment and determine ecologic stability. The trees damaged by radionuclides lose their environmental characteristics (Butkus *et al.* 2007).

The growth of annual tree rings provides information on the pollution of the area and they are a distinctive indicative figure for ecological condition of the environment.

The extend of radionuclide accumulations and migrations in trees depend on the density of a plant canopy, a type of a plant, climate conditions, and physical and chemical composition of soil (Pederson *et. al.* 2004; Butkus *et. al.* 2004). The accumulation of radionuclides in plants may depend on the scenery and plant life, the litterfall and characteristics of soil, also on the extent of radionuclide pollution, their chemical characteristics and meteorological conditions (Papastefanou *et. al.* 1999).

A near surface repository for low and intermediate-level short-lived radioactive waste is being constructed on Stabatiškės site near to Ignalina Nuclear Power Plant. In order to measure potential migration and distribution of radionuclides using the near surface repository for radioactive waste, it is necessary to know the existing background activity of radionuclides in the surroundings of Stabatiškės site.

It was decided to assess specific activity of radionuclides in scots pine wood of Stabatiškės site. Distinct concentric rings or annual rings which allow investigation on radionuclide pollution in a particular area during different periods of time are clearly visible in the cross-section of softwood stem. While growing, annual rings store information about ongoing processes in the environment and in this way become nature monitors (Stravinskienė 2001).

The aim of this work is to assess the dynamics of radionuclide distribution in the wood of the Scots pine (*Pinus sylvestris* L.) which is a representative of coniferous trees dominant in the area of Stabatiškės site.

2. Methodology

Samples of the Scots pine wood (*Pinus sylvestris* L.) were taken in the western area of Stabatiškės site (coordinates by VMS-1994: X=6163652, Y=656459), 1 km away from the second reactor of Ignalina Nuclear Power Plant (Fig. 1).

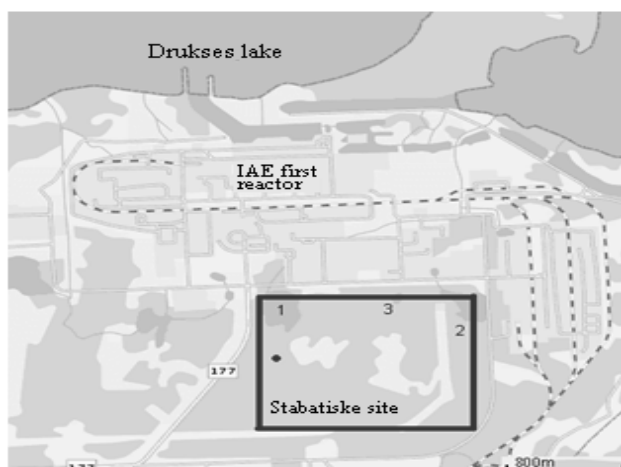


Fig 1. Stabatiškės site: ●—area where the wood samples were taken from; □—area of Stabatiškės site; 1, 2 —water (a pond); 3—marshy land

During the construction of Ignalina NPP the former village of Stabatiškės was polluted with construction waste; the natural water flow was unbalanced and the soil layer was affected. Trees and bushes occupy the greater part of the area of the former Stabatiškės village (Radioaktyviosios atliekos 2006).

Stabatiškės site consists of a loam and sandy loam hill with bushes and trees growing on it. In the middle part of the site there are two oblong hills with marshy valleys surrounding them. Two water bodies and a marshy area are found in the northern part of the site. In the western part of the site there is a road leading from Ignalina NPP to Visaginas (Fig. 1).

As the stem comprises the main part of a pine (*Pinus sylvestris* L.) biomass (about 80 %) (Butkus and Pliopaitė Bataitienė 2006), the sample to measure the specific activity of radionuclides was taken from the stem.

The stem sample is a wood roll which diameter is equal to the stem diameter and its height is $h=3-5$ cm.

The wood roll was taken from the lower part of the stem (1–1.5 m above the litterfall). Using chisels the stem roll was split according to years every three rings. Also using chisels was split and bark.

The finished wood samples were weighed (weight = 0.003–0.153 kg) and dried to constant mass at room temperature (17–22 °C). The dried stem samples were burnt at 480 °C temperature for 2 hours in a muffle furnace. The charcoal was then weighed, crushed with a pestle, poured into piles and the specific activities of radionuclides in charcoal were measured using a gamma spectrometer (Ge(Li)) with a semiconductor detector.

The Ge (Li) semiconductor spectrometer was used to determine the specific activities of radionuclides.

The study samples were examined in “Denta” cuvettes for 1–2 days.

Using the semiconductor spectrometer, the activities of these radionuclides: ^{226}Ra , ^{214}Pb , ^{208}Tl , ^{214}Bi , ^{137}Cs , ^{227}Ac , ^{40}K , ^{232}Th , were measured in the samples.

Radionuclides are identified according to radiation energy characteristic to each radionuclide: ^{214}Pb – (295 keV, 352 keV); ^{208}Tl – 583 keV; ^{214}Bi – 609 keV; ^{137}Cs – 662 keV; ^{227}Ac – 911 keV, 969 keV; ^{40}K – 1460 keV; ^{60}Co – 1173 keV.

The specific activity of ^{226}Ra is determined by calculating average specific activity of ^{214}Pb (295 keV, 352 keV) and ^{214}Bi (609 keV). The specific activity of ^{232}Th is determined by calculating the average specific activity of ^{228}Ac (911 keV) and ^{208}Tl (583 keV).

Based on the radionuclide activity in the sample, its specific (Bq/kg) or volumic (Bq/m³) activity is calculated.

The specific activity of radionuclides is determined by the formula (LAND 36–2000):

$$A_a = \frac{\frac{S}{t} - \frac{S_f}{t_f}}{\eta \cdot \varepsilon \cdot m}, \quad (1)$$

here: A_a – specific activity of the examined radionuclide in the sample, Bq/kg; S – radionuclide peak area obtained while measuring radionuclide activity in the sample, imp; t – duration of a sample measurement, s; S_f – peak area obtained while measuring background radiation, imp; t_f – duration of background radiation measurement, s; η – quantum yield of radionuclide decay energy; ε – efficiency of the spectrometric system; m – the sample weight, kg.

The specific activity of radionuclides in the stem charcoal is recalculated for natural weight of the stem wood:

$$A_{nat} = \frac{m_a \cdot A_a}{m_{\xi}}, \text{ kai } m_{a.v.} = m_a \quad (2)$$

$$A_{nat} = \frac{m_a \cdot A_a}{m_{\xi}} \cdot \frac{m_a}{m_{a.v.}}, \text{ kai } m_{a.v.} > m_a \quad (3)$$

here: A_{nat} – specific activity of radionuclides in natural wood in 2010, Bq/kg; m_a – charcoal weight in measurement cuvettes, kg; A_a – specific activity of radionuclides in a sample charcoal, Bq/kg; m_{ξ} – natural weight of sample wood, kg; $m_{a.v.}$ – total weight of sample charcoals, kg.

The specific activity of radionuclides which was in wood during the year of the ring formation is calculated using the formula:

$$A_r = A_{nat} \cdot e^{\lambda \cdot t}, \quad (4)$$

here: A_r – specific activity of radionuclides during the period of the ring formation, Bq/kg; t – time interval (from the ring formation to 2010) during which the decline of activity is examined, y.; λ – radioactive decay constant, y^{-1} .

The absolute error of specific activity is calculated using the formula:

$$\Delta A = A \left(\frac{p}{100} + \frac{\Delta t}{t} + \frac{\Delta m}{m} \right), \quad (5)$$

here: A – specific activity of the examined radionuclide in the sample which was determined by the first formula, Bq/kg; p – relative error of a measurement estimated by the spectrometer, %; Δt – error of measurement duration, s; Δm – error of the sample weight measurement, kg.

The population correlation coefficient $\rho_{x,y}$ between two random variables X or Y with expected values μ_x and μ_y and standard deviation σ_x and σ_y is defined as:

$$\rho_{x,y} = \text{corr}(X, Y) = \frac{\text{cov}(X, Y)}{\sigma_x \sigma_y} = \frac{E[(X - \mu_x)(Y - \mu_y)]}{\sigma_x \sigma_y}, \quad (6)$$

here: E is the expected value operator, cov means covariance, and corr a widely used alternative notation for Pearson's correlation.

3. Results and their analysis

The stem samples of Scots pinewood (*Pinus sylvestris* L.) were made in segments. The stem segment is a part of the stem sample consisting of 3 annual rings of the pine. Sample consisting 3 annual rings of the pine, because it is very difficult to identify specific activity of radionuclides if sample weight is too low.

Figure 2 shows how the biomass of the stem of Scots pinewood (*Pinus sylvestris* L.) which was taken 1 m above from the litterfall is changing. This change of biomass was experimentally determined.

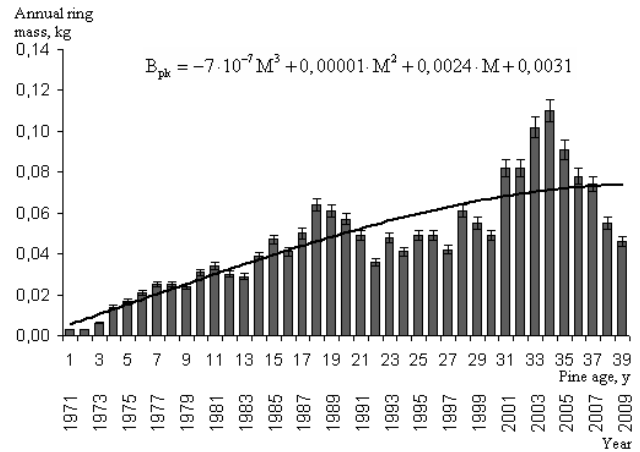


Fig 2. The change of Scots pine biomass 1971–2009

The biomass change can be described by the fourth-order polynomial equation:

$$B_{pk} = -7 \cdot 10^{-7} \cdot M^3 + 0.00001 \cdot M^2 + 0.0024 \cdot M + 0.0031 \quad (7)$$

here: B_{pk} – biomass of a segment of an annual ring, kg; M – age of the pine, y.

The dendrochronological and dendroindicational research determined that the width and weight of annual tree rings depend on anthropogenic environmental factors and climate change (Stravinskienė 2002). In their works Bitvinskis (1997) as well as Stravinskienė and Erlickytė-Marčiukaitienė (2009) highlights that the peculiarities of the dynamics of annual radial growth of trees are caused by biological characteristics of tree species, ecological conditions of the place where a plant grows, and the long-term change of climate factors.

The data in Fig. 2 shows that there is no linear dependence between the change of wood biomass and the age of wood. Such a change in the speed of the biomass growth might have been caused by favourable and unfavourable climatic conditions and atmospheric pollution.

During the 39-year growth period the pine biomass was changing unevenly. Previous researches (Bitvinskis 1989; Stravinskienė *et al.* 1981) showed that in Lithuanian forests the maxima and minima of annual radial growth of conifer trees manifest themselves on specific phases of 22-year cycle of solar activity. The larger is the

amplitude of solar activity, the higher are the oscillation amplitudes of annual radial growth of conifer trees.

Buitkuvienė (1998) identified the decline period of annual radial growth of conifer trees which started since 1990 and lasted till 1996. This coincides with the growth period of the studied pine when it was between 20 and 30 years old. During those years the decline in the pine radial growth was influenced by unfavourable climatic conditions and environmental pollution.

The analysis of the data from Fig. 2 shows the decline in wood biomass growth in 2005–2010. This decline in wood biomass growth might have been caused by climate change or by some pine tree disease.

According to the methodology described in the methodology chapter, radiometric analysis determined the specific activities of artificial (^{137}Cs) and natural (^{226}Ra , ^{232}Th , ^{40}K) radionuclides in the samples of the pinewood taken from Stabatiškės site.

Fig. 3 shows the specific activities of natural ^{226}Ra and ^{232}Th radionuclides.

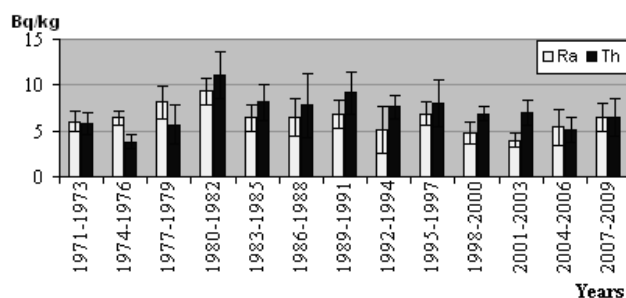


Fig 3. The special activities of ^{226}Ra , ^{232}Th in the pinewood

The special activities of ^{226}Ra in the annual tree rings change from $4,0 \pm 0,7$ to $9,3 \pm 1,4$ Bq/kg, and the specific activities of ^{232}Th vary from $3,8 \pm 0,8$ to $11,1 \pm 2,6$ Bq/kg. The biggest values of the special activities of ^{226}Ra and ^{232}Th were found during 1980–1982 pine growth years.

In conclusions, the distribution of natural radionuclides in the sample wood suggests that the radionuclides ^{226}Ra and ^{232}Th might get into a plant influenced by the physical and chemical characteristics of soil and meteorological conditions.

The specific activities of ^{226}Ra and ^{232}Th in pinewood increase between the years 1971 and 1982 and then start decreasing. This might be related to the changes in the biomass growth of tree rings because the data in Fig. 2 shows that the biomass growth of the Scots pine tree increases linearly in 1971–1982 and since 1982 the changes of the biomass growth become uneven.

Fig. 4 and Fig. 5 present curves showing the relationship between the biomass change of pine rings and the specific activities of ^{226}Ra and ^{232}Th .

The data in Fig. 4 and 5 indicates that the 1971–1982 increase the growth of wood biomass increment increased specific activity of ^{226}Ra and ^{232}Th of pine rings.

The relationship between the specific activities of radionuclides and the growth of the ring biomass is expressed in the formulas which can be seen in Fig. 4 and Fig. 5.

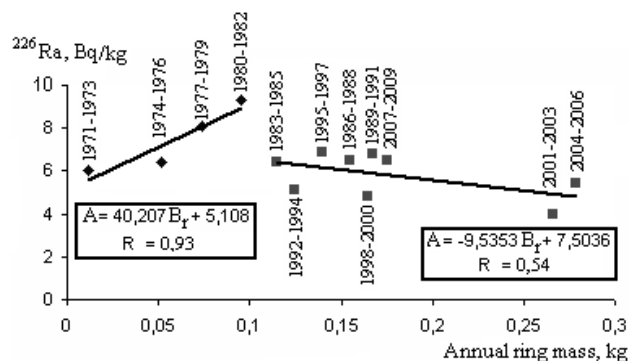


Fig 4. The relationship between the wood biomass and the specific activity of ^{226}Ra

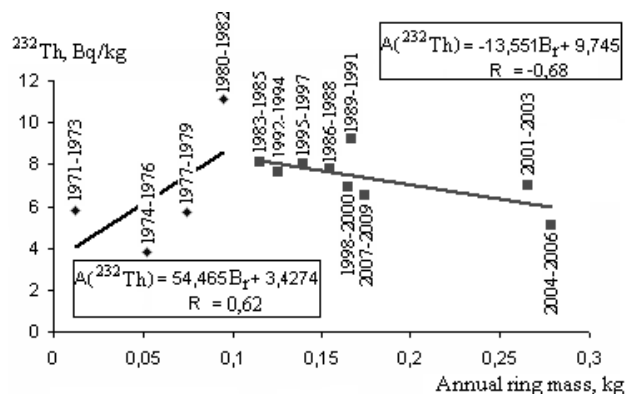


Fig 5. The relationship between the wood biomass and the specific activity of ^{232}Th

After summarizing the data in Fig. 4 and 5, it was understood that the pine absorbed radionuclides ^{226}Ra and ^{232}Th most efficiently in 1971–1982. This corresponds to the period of pine growth when it was 1–12 years old.

Fig. 6 presents the measurement results of the special activities of ^{40}K in pinewood.

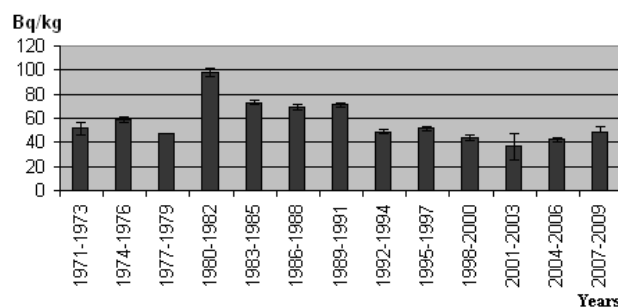


Fig 6. The change of the specific activities of ^{40}K in the pinewood

As the data in Fig. 6 shows, in 1971–1979 the specific activity of ^{40}K in the pinewood was stable but since 1980 the specific activity of ^{40}K started decreasing depending on the age of the wood. The specific activities of ^{40}K in annual tree rings change from $34,4 \pm 11,1$ to $70,0 \pm 4,9$ Bq/kg. The greatest specific activity of radionuclides was found in 1980–1982. Most probably it was caused by the sudden decrease in the biomass growth which was characteristic to this period (Fig. 2).

Fig. 7 shows the dependence of the growth of the ring biomass and the specific activities of ^{40}K in annual tree rings.

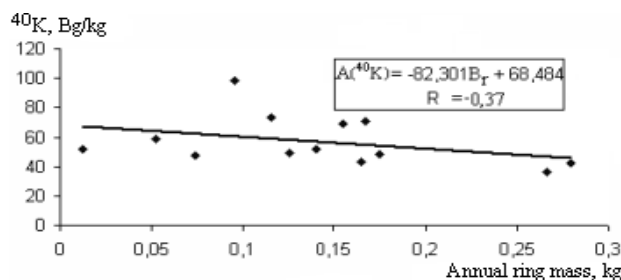


Fig 7. The relationship between the wood biomass and the specific activity of ^{40}K

The data in Fig. 7 indicates that the specific activity of ^{40}K in annual tree rings and the growth of the wood biomass are in inverse proportion.

Fig. 8 presents the change in specific activities of the artificial radionuclide ^{137}Cs in pinewood.

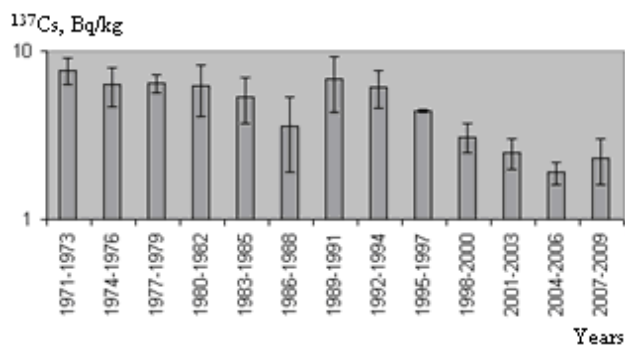


Fig 8. The specific activities of ^{137}Cs in the pinewood

As the data in Fig. 8 shows, the activities of ^{137}Cs in the annual tree rings change from 1.9 ± 0.3 to 7.7 ± 1.4 Bq/kg. The greatest specific activity of the radionuclide was measured at the stage of wood formation.

Butkus and Pliopaitė Bataitienė (2006) determined that the change of the specific activity of ^{137}Cs in wood depends on the speed of its biomass growth. Fig. 9 presents the dependence of the growth of the wood biomass and the specific activities of ^{137}Cs in individual tree rings.

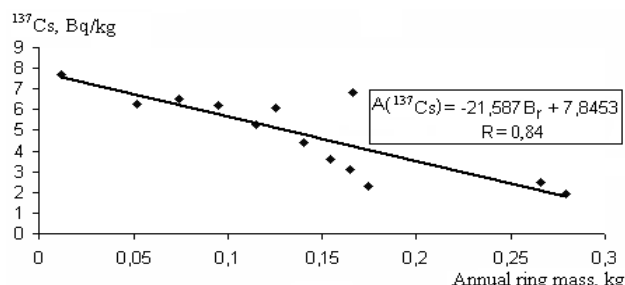


Fig 9. The relationship between the wood biomass and the specific activity of ^{137}Cs

The analysis of the data from Fig. 9 shows that the special activity of ^{137}Cs is bigger when the growth of the wood biomass is smaller. As the correlation coefficient

between the growth of the wood biomass and the specific activity of ^{137}Cs is $R = -0,84$, it suggests that there is a strong reverse relationship between these two values, i.e., the bigger is the biomass growth, the smaller is the specific activity of ^{137}Cs .

Fig. 10 shows the specific activity of radionuclides in the bark of the pine.

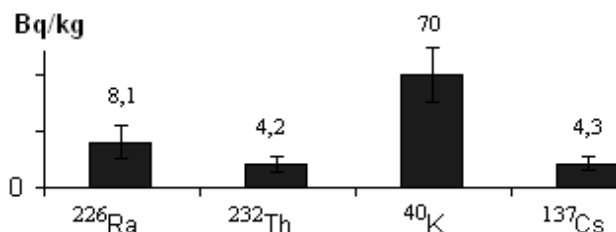


Fig 10. The values of average specific activities of radionuclides in the bark of the pine

Beinaravičius determined (2005) that there are twice as many artificial radionuclides in the bark as in the wood itself and the bark contains several times more ^{40}K as well. However, according to the data in fig. 10, the specific activity of ^{137}Cs in the pine bark corresponds to the value of the average radionuclide activity in the whole wood.

The specific activity of ^{40}K in the bark is 70.0 ± 4.9 Bq/kg. Regardless of the biggest specific activity of radionuclides which was determined in 1980–1982 during the wood formation period, the specific activity of ^{40}K in the bark is equal to the average specific activity in the wood samples.

The specific activities of ^{226}Ra and ^{232}Th in the bark differ little from their value in the wood samples and they are respectively equal to 8.1 ± 2.9 or 4.2 ± 1.3 Bq/kg.

Such values of specific activities of radionuclides in the bark of pinewood might depend on the density of the crown of the plant and on climatic conditions. The bark separated from the stem had a largest mass of all the segments of the trunk. This might be related to plenty of precipitation in the territory of Stabatiškės site.

Conclusions

1. The decrease of the speed of the pine biomass growth is observed in 1990–1996 and 2005–2010. Unfavourable climatic conditions caused the reduction in radial growth.
2. The specific activities of ^{226}Ra in annual tree rings change from 4.0 ± 0.7 to 9.3 ± 1.4 Bq/kg, and the special activities of ^{232}Th vary from 3.8 ± 0.8 to 11.1 ± 2.6 Bq/kg. The biggest values of the special activities of ^{226}Ra and ^{232}Th were found during 1980–1982 pine growth years. This corresponds to the period of pine growth when it was 10–12 years old.
3. In 1971–1982 when the growth of ring biomass started increasing, the specific activities of ^{226}Ra and ^{232}Th in the annual rings were also growing but since 1982 the special activities of radionuclides started decreasing.
4. The special activity of ^{40}K in the pinewood was stable but since 1980 it started decreasing depending on the

age of the wood. The specific activities of ^{40}K in annual tree rings change from 34.4 ± 11.1 to 70.0 ± 4.9 Bq/kg. The special activity of ^{40}K in annual tree rings and the growth of wood biomass are in reverse proportion.

5. The special activities of ^{137}Cs in the annual tree rings change from 1.9 ± 0.3 iki 7.7 ± 1.4 Bq/kg. The greatest specific activity of the radionuclide was measured at the stage of wood formation, in 1971–1973.
6. As the correlation coefficient between the growth of the wood biomass and the specific activity of ^{137}Cs is $R = -0.84$, it suggests that there is a strong reverse relationship between these two values, i.e., the bigger is the biomass growth, the smaller is the specific activity of ^{137}Cs .
7. The specific activity of ^{137}Cs in the pine bark corresponds to the value of the average radionuclide activity in the whole wood. Regardless of the biggest specific activity of ^{40}K , the specific activity of this radionuclide, as well as the specific activities of ^{226}Ra and ^{232}Th , in the bark differs little from their value in the wood samples.

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