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EVALUATION OF ^{137}Cs AND ^{90}Sr TRANSFER FROM SOIL TO SCOTS PINE (*PINUS SYLVESTRIS* L.) BY THEIR DISCRIMINATION COEFFICIENTS

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Abstract. Radionuclides migrate in the soil in both vertical and horizontal directions. Radionuclide migration is predetermined by physical and chemical properties, climatic conditions, environmental relief, soil type, hydrological regime in the territory, the type of flora, agrochemical specificities of agriculture and other factors. While making an analysis of ^{137}Cs behaviour in the forest ecosystem, two processes are distinguished, as a result of which ^{137}Cs enters a tree: due to fallout on the top and trunk of a tree and because of cesium assimilation through roots from the soil. It should be mentioned that internal change of cesium in a tree also occurs; however, it is not as distinct as cesium uptake from the atmosphere and soil. ^{90}Sr, having deposited from the atmosphere on the soil, under the effect of natural factors, participates in the migration process of substances.

With account taken of the physiological specific features of the plant of each species, plants assimilate certain ions from the soil through the root system. Of monovalent ions, ^{40}K is the most important for the plants. In the case of potassium shortage, plants assimilate other monovalent ions, like cesium. This phenomenon is called the potassium discrimination effect on cesium transfer. The discrimination coefficient is estimated according to ^{137}Cs and ^{40}K activities in the soil and the plant. It is assumed that potassium ions may predetermine cesium assimilation. This expression of discrimination coefficient may be also applied for ^{90}Sr, but in this case instead of ^{40}K specific activities Ca is evaluated, and instead of ^{137}Cs – ^{90}Sr. The discrimination effect of ^{90}Sr on assimilation depends greatly on the plant species. The accumulated amount of ^{90}Sr in the plant depends also on the presence of the proper forms of calcium in the soil solution, close to the root area.

Keywords: ^{137}Cs, ^{90}Sr, radionuclide transfer, radionuclide chemical analogue, discrimination coefficient.

1. Introduction

One of the main contamination sources of artificial ionizing radiation is fallout after nuclear weapon tests, accidents at nuclear power plants, nuclear fuel processing enterprises and operating nuclear power plants. Some part of radionuclides gets into plants directly; another part enters the soil and its waters (Chad-Umoren and Briggs-Kamara 2010). Radionuclides with atmospheric fallouts through the aboveground part and with the soil solution through the root system enter plants and get redistributed inside them. The character of radionuclide redistribution depends on the level of contamination, the prevailing soil type, chemical and physical properties of radionuclide, the chemical form of radionuclide, the plant species, and the climatic conditions (Щеглов and Цветнова 2004).

^{137}Cs and ^{90}Sr are attributed to radionuclides of artificial origin, which emerged in the environment due to human activities. These radionuclides are noted for their negative impact on the environment and its components. ^{90}Sr is β emitter, and ^{137}Cs is β emitter, which is identified according to the γ radiation emitted by the decay product ^{137m}Ba. The main hazard to living organisms is related to ionizing radiation of radionuclides which entered the organism, its distribution in the organism and the resulting irradiance (Cesium 2006; Strontium-90 2006; Čepanko *et al.* 2006).

In the work (Sanches *et al.* 2008), it is stated that ^{137}Cs is noted for high mobility in the plant, and the higher values of its specific activity are determined in the growing parts of the plant, for example, in the bark and twigs. The work authors (Barci-Funel *et al.* 1995), while studying conifers (a pine (*Pinus silvestris*), a picea (*Pinus picea*), a larch (*Lari*x)), identified that the highest ^{137}Cs specific activities are in their needles, twigs, and bark. In these wood components ^{137}Cs specific activity was determined higher than in the wood of big branches. In the bark ^{137}Cs specific activity is by several rows higher than in the inner parts of wood components. Similar data are also provided in the work (McGee *et al.* 2000), but these researchers determined the smaller difference of ^{137}Cs specific activity between the external and internal parts of the component, only five times. The authors of the work (Fogh and Anderson 2001) specify this difference as the differences of specific activities in young and old parts of wood components, in young parts of the component ^{137}Cs specific activity is by one row higher than in old ones.

Chemical analogue of ^{137}Cs is ^{40}K. Potassium is an important macroelement for plants; it is responsible for the correction of the osmotic pressure in a cell and predetermines the plant growth. Some plants may assimilate the big amount of K ions with Na ions, not experiencing any negative effect on plant growth. Meanwhile, additional growth may be stimulated for other plants, resulting in the increment of K^+ in a plant. Growth stimulation with Na ions is promoted due to its ability to expand a cell and because of the water balance in a plant. In the scientific literature it is mentioned that Na, K, Cs ions may rival, compete, when entering the plant, and it is also stated that ^{137}Cs may be used as Na carrier in the plant nutrition chain (Marschner 1995). With consideration of the physiological specificities of each species of a plant, plants assimilate certain ions from the soil through the system of roots. Of monovalent ions, ^{40}K is the most important for plants. In the case of potassium shortage, plants assimilate other monovalent ions, like cesium. This phenomenon is referred to as a potassium discrimination effect on cesium transfer (Solecki and Chibowski 2002).

^{90}Sr is an artificial radionuclide, which is one of the most essential in terms of radioecology, and is spread in the terrestrial ecosystem (Kliashtorin *et al.* 1994; Willey and Fawcett 2006). It is available in the soil solution in almost all mineral soils, contaminated with it (Frissel 1992). This radionuclide transfer in the soil is fast (Kliashtorin *et al.* 1994), and transfer to plants is high (Ban-nai and Muramatsu 2002). ^{90}Sr transfer in the soil-to-plant system depends greatly on the soil properties (Roca *et al.* 1997; Askbrant and Sandalls 1998), but the plant species is also of no less importance (Veresoglou *et al.* 1995).

The chemical analogue of ^{90}Sr radionuclide is Ca. The latter gets accumulated in microorganisms, plants and animals, performing the principal physiological functions (White and Broadley 2003). Ca is a macroelement, important to plants, which enters plants from the soil solution to shoots (White 2001). Ca and ^{90}Sr are competing elements in the soil-to-plant transfer (Veresoglou *et al.* 1995).

Competitive factors may be evaluated by discrimination coefficients, which are estimated according to ^{137}Cs and ^{40}K activities in the soil and the plant. According to research, conducted by the work authors (Solecki and Chibowski 2002), a conclusion was made that potassium ions may greatly predetermine cesium assimilation. This discrimination coefficient expression may be also applied for ^{90}Sr, but in this case instead of ^{40}K specific activities Ca is evaluated, and instead of ^{137}Cs – ^{90}Sr. The authors state that the discrimination effect of Ca assimilation on ^{90}Sr depends strongly on the plant species. The accumulated amount of ^{90}Sr in the plant also depends on the existence of the adequate forms of calcium in the soil solution, close to the root zone of the plant (Solecki and Chibowski 2002).

According to the results, obtained by the authors of the work (Solecki and Chibowski 2002), it is possible to say that ^{137}Cs soil-to-plant transfer coefficients in many explored cases are approximately by two times higher than for ^{90}Sr. Meanwhile, a ^{137}Cs discrimination effect on ^{40}K assimilation in grasses is established as being higher than ^{90}Sr discrimination effects on Ca assimilation.

The purpose of this work is according to the determined discrimination coefficients to analyze the competitiveness of artificial (^{137}Cs and ^{90}Sr) radionuclides and their chemical analogues (^{40}K and Ca accordingly) in entering the plant.

2. Methods for determination of discrimination coefficients

A Scots pine under study grew in the Neris Regional Park in the vicinity of the Paaliosė village. Prior to the accident at the Chernobyl Nuclear Power Plant (ChNPP), ^{137}Cs radioactive contamination in this locality was from 7 Bq/kg to 12 Bq/kg. After the ChNPP accident, ^{137}Cs radioactive contamination on the Paaliosė growing site increased (3700 – 7400 Bq/m^2). ^{90}Sr contamination after the ChNPP accident varies in the natural landscape and the soil. In this locality, it changed from 550 to 1300 Bq/m^2 in the soils, and in the natural landscape on the territories of the growing sites or close to them to 300 Bq/m^2 (Butkus *et al.* 1999).

Discrimination coefficients were determined for Scots pine (*Pinus sylvestris* L.) components and annual rings. A discrimination coefficient is estimated according to ^{137}Cs and ^{40}K activities in the soil and the plant by the following expression (Solecki and Chibowski 2002):

$$K_{\mathrm{d},^{137}\mathrm{Cs}} = \frac{\left(\dfrac{A_{^{137}\mathrm{Cs}}}{A_{^{40}\mathrm{K}}}\right)_{\mathrm{plant}}}{\left(\dfrac{A_{^{137}\mathrm{Cs}}}{A_{^{40}\mathrm{K}}}\right)_{\mathrm{soil}}} = \frac{PK_{^{137}\mathrm{Cs}}}{PK_{^{40}\mathrm{K}}}, \qquad (1)$$

where: $K_{\mathrm{d},^{137}\mathrm{Cs}}$ – discrimination coefficient; $A_{^{137}\mathrm{Cs}}$, $A_{^{40}K}$ – ^{137}Cs and ^{40}K specific activities in the plant and the soil, Bq/kq; $PK_{^{137}\mathrm{Cs}}$, $PK_{^{40}\mathrm{K}}$ ^{137}Cs and ^{40}K soil-to-plant transfer coefficient, which may be determined according to the provided methods (Pliopaitė Bataitienė and Butkus 2008).

Discrimination coefficients absolute error is calculated by the formula (Баранов *et al.* 1966):

$$\Delta K_{\mathrm{d}} = \overline{K}_d \sqrt{\left(\frac{\Delta A_m}{\overline{A}_m}\right)^2 + \left(\frac{\Delta A_d}{\overline{A}_d}\right)^2}, \qquad (2)$$

where: ΔK_d – discrimination coefficients error, m^2/kg; $\overline{K}_d$ – average of discrimination coefficients, m^2/kg; ΔA_{m} – error of radionuclide specific activity in wood, Bq/kg; $\overline{A}_m$ – average of radionuclides specific activity in wood, Bq/kg; ΔA_{d} – error of radionuclide activity in soil, Bq/m^2; $\overline{A}_d$ - average of radionuclide activity in soil, Bq/m^2.

The ^{90}Sr discrimination effect on Ca assimilation depends greatly on the plant species. The accumulated amount of ^{90}Sr in the plant depends on the presence of adequate forms of calcium in the soil solution, close to the zone of the plant roots (Solecki and Chibowski 2002). The ^{90}Sr discrimination coefficient is evaluated by recalculating the ^{90}Sr concentration and Ca specific activity into the number of atoms of those elements.

^{137}Cs and ^{40}K specific activities are measured with a semiconductor Ge-Li gamma spectrometer. Specific activities were determined according to the formula (Land 36 – 2000; Butkus *et al.* 2008):

$$A = \frac{\frac{S}{t_1} - \frac{S_f}{t_f}}{\eta \cdot \varepsilon \cdot m}, \tag{3}$$

where: A – radionuclide specific activity in the sample (Bq/kg); S – radionuclide peak area, received by measuring radionuclide activities in the sample, imp.; S_f – radionuclide peak area, received by measuring the activity of background radiation of radionuclides, imp.; t_1 – measuring time of radionuclide activity in the sample, s; t_f – measuring time of background radiation activity of radionuclide, s; η - quantum energy output of radionuclide radiation; ε – spectrometer efficiency; m – sample weight, kg.

Specific activity absolute error is calculated by means of the formula:

$$\Delta A = A\left(\frac{p}{100} + \frac{\Delta t}{t} + \frac{\Delta m}{m}\right), \tag{4}$$

where: A – radionuclide specific activity in the sample, calculated by the formula (2), Bq/kg; p – relative error of measurement, determined by spectrometer, %; Δt – measuring time error, s; Δm – sample weight determination error, kg.

^{90}Sr specific activity is determined by the low background beta radiometer UMF – 1500M. Specific activity is calculated by means of the formula (Land 64-2005):

$$A_{Sr-90} = \frac{A}{Y \cdot m}, \tag{5}$$

where: A_{Sr-90} – ^{90}Sr specific activity in the sample, Bq/kg ; A – activity in the sample, Bq; Y – ^{90}Y chemical output, %; m – sample weight, kg.

Standard error of the sample activity is calculated by means of the formula (Land 64-2005):

$$Sc = \frac{\sqrt{\frac{N}{t} + \frac{N_f}{t_f}}}{E \cdot \exp(-\lambda \Delta t)}, \tag{6}$$

where: Sc – standard activity deviation, Bq/sample.; N – the intensity of sample impulses, imp./s; N_f – background impulse calculation speed, imp./s; t –impulse calculation time, s; t_f – the intensity of background radiation impulses; E – registration efficiency, imp./s/Bq; λ – ^{90}Y transformation constant, 0,0108 h^{-1}; Δt – time interval between ^{90}Y isolation and measuring time, h.

For Ca investigation, samples are incinerated in the muffle furnace to ashes at a temperature of 450°C. Ashes of the sample are dissolved in the solution, for the preparation of which HNO_3 concentrated is needed (per sample 10 ml HNO_3 concentrated and 2ml H_2O_2). Approximately 0,25g sample ashes are taken for investigation. Samples, in the above-mentioned solution, are dissolved in the mineralizer. Later, after the solutions got cooled, nuclear absorption spectrum analysis is carried out, applying the nuclear absorption spectrophotometer 210 VGP Buck Scientific (with the flame), during which Ca concentration in the sample ashes is determined:

$$C(Ca, p) = \frac{C^* \cdot K_{Sk} \cdot V}{m_p}, \tag{7}$$

where: $C_{(Ca,p)}$ – Ca concentration in ashes, mg/kg; C^* – Ca concentration in the sample extract, mg/l; K_{sk} – dilution coefficient, V – the prepared sample volume, ml; m_p – ash mass taken for investigation, g.

Ca amount in the sample is recalculated by means of the formula:

$$C(Ca,b) = C(Ca,p) \cdot \left(1 - \frac{m_{befor} - m_{after}}{m_{befor}}\right), \tag{8}$$

where: $C_{(Ca,p)}$ – Ca concentration in ashes, mg/kg; m_{befor} – raw mass of the sample, kg; m_{after}– burnt sample mass, kg.

3. Investigation results of ^{137}Cs and ^{90}Sr discrimination coefficients

^{137}Cs and ^{90}Sr discrimination coefficients, describing the specific features of the entrance of artificial radionuclides (^{137}Cs and ^{90}Sr) and their chemical analogues (^{40}K and Ca) from the soil to Scots pine (*Pinus sylvestris* L.) components, are presented in Fig 1.

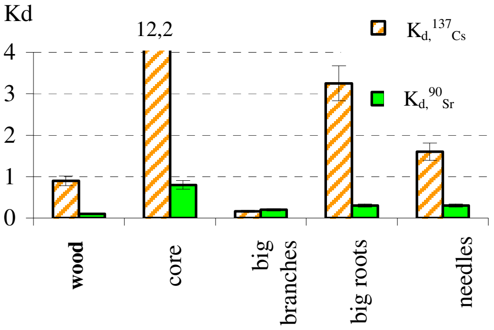

Fig 1. ^{137}Cs and ^{90}Sr discrimination coefficients for Scots pine components of the Paaliosė growing site

Highest ^{137}Cs discrimination coefficients for Scots pine bark (12,2) and big roots (10,5) are determined. As already mentioned, the discrimination coefficient specifies the entrance of radionuclide and its chemical analogue (in the case under study, ^{137}Cs and ^{40}K) from the soil to the plant. The obtained discrimination coefficient results for big roots confirm that in this pine component for ^{137}Cs distribution ^{137}Cs and ^{40}K ratio in the soil is of special importance. However, the process of ^{137}Cs transfer into the bark is predetermined not only by the transfer from the soil; here a considerable contribution also belongs to the entrance of the radionuclide from the atmosphere. Therefore, it is probable that the received ^{137}Cs discrimination coefficient for Scots pine bark is highest due to the ^{137}Cs entrance from the atmosphere and soil, and the received discrimination coefficient not fully reflects the effect of ^{137}Cs on the entrance of ^{140}K from the soil to the bark.

The highest values of ^{90}Sr discrimination coefficients were determined for Scots pine bark, needles and roots – i.e. those tree components where metabolism is more intensive. For some components, bark and needles, in addition to the entrance of radionuclides from the soil with nutrient materials, another pathway for radionuclide entrance is from the atmosphere; probably, therefore, the higher values of ^{90}Sr discrimination coefficients are received.

To evaluate the change of many years of ^{137}Cs discrimination coefficient in tree rings, it was accepted that:

1. ^{137}Cs distribution in certain rings is restored by year after the evaluation of radioactive decay, and no account is taken of the radical movement of the radionuclide towards the core.
2. ^{137}Cs distribution in the soil is taken at the depth of 0–25 cm, since here the main mass of Scots pine roots is located. Distribution of this radionuclide in the soil in the corresponding year is restored by applying diffusion equation decision, to be used for the evaluation of radionuclide dispersion in the soil.
3. ^{40}K distribution in the soil is even and not subject to change within Scots pine growth period.

Fig 2 presents ^{137}Cs discrimination coefficient long-term change in Scots pine annual rings in the Paaliosė growing site.

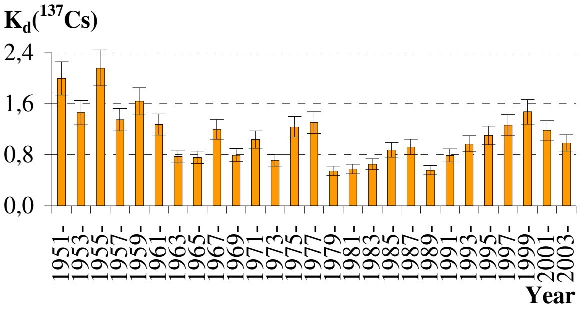

Fig 2. ^{137}Cs discrimination coefficient change of many years in the Scots pine annual rings in the Paaliosė growing site

In the change of many years of ^{137}Cs discrimination coefficients in Scots pine rings, it is possible to distinguish two periods when discrimination coefficients are higher than average, i.e. from 1951 to 1962 and from 1991 to 2004. Within those periods, ^{137}Cs soil-to-wood transfer is increased. Such long-term distribution of discrimination coefficients could be predetermined by radioactive contamination of the soil with cesium. From 1991 to 2004 cesium soil-to-wood transfers may be related to the possibility of the secondary ChNPP contamination through the tree root system, i.e. from 1951 to 1962 cesium soil-to-wood transfer was predetermined by nuclear explosions. The consequences of this radioactive soil contamination, however, should appear approximately from the year 1955. The results obtained within this period may be related to ^{137}Cs radial mobility towards the tree core; in addition, with the more intensive metabolism of a young tree and with the better possibility for ^{137}Cs to enter wood.

Fig 3 provides ^{90}Sr discrimination coefficient many-year change in the wood.

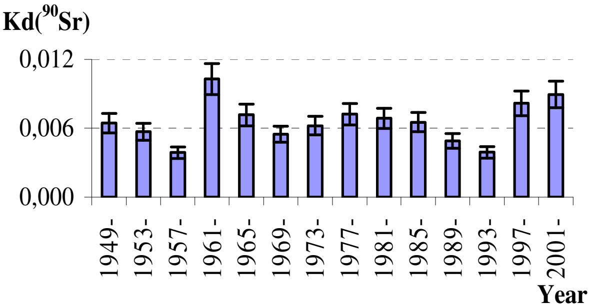

Fig 3. ^{90}Sr discrimination coefficient change of many years in Scots pine wood on the Paaliosė growing site

Change in ^{90}Sr discrimination coefficients is even, i.e. no considerable increments of ^{90}Sr soil-to-wood transfer exist, even though within the periods of 1961–1968, 1973–1988, 1997–2004, the values of ^{90}Sr discrimination coefficient, higher than average, were determined, which may be explained by the ^{90}Sr contamination as a result of nuclear explosions (within the periods of 1961–1968, 1973–1988) that reached such depth of the soil where a large part of the biomass of pine roots is located, and in 1997–2004 the contamination from the ChNPP accident, which was insignificant in the territory of Lithuania – only by 1.1 times higher than before the ChNPP accident, reached such depth where the big part of the biomass of pine roots is located.

Comparing ^{137}Cs and ^{90}Sr discrimination coefficients, it was determined that ^{137}Cs discrimination coefficients, on the average, are by two rows higher than ^{90}Sr discrimination coefficients. Different distribution of discrimination coefficients in annual rings, which are closer to the core, was noticed. ^{137}Cs discrimination coefficients tend to increase towards the core and is by 1.2–1.9 times higher than the average ($K_{d,av.}$ = 1.1), and ^{90}Sr remains close to the average ($K_{d,av.}$ = 0.006). Such distribution of discrimination coefficients in the rings, existing closer to the core, confirms the thought that ^{137}Cs in the gymnosperms, whereto the conifers are attributed, is radially mobile, and the mobility of ^{90}Sr is so small that it may be treated as stable in the conifers.

4. Conclusions

1. The highest ^{137}Cs and ^{90}Sr discrimination coefficients were determined needles and the roots.
2. ^{137}Cs and ^{90}Sr discrimination coefficients, in evaluating the transfer of ^{137}Cs and ^{90}Sr from the soil to Scots pine wood, differs by two rows.

3. ^{137}Cs discrimination coefficients in wood rings, existing close to the core, increases and is by 1.2–1.9 times higher than the average ^{137}Cs discrimination coefficient ($K_{d,av.} = 1.1$).
4. ^{90}Sr coefficients in wood rings, existing close to the core, remain close to the average (0.006).
5. The specific features of the distribution of ^{137}Cs and ^{90}Sr discrimination coefficients in the rings, existing closer to the core, reflect the intensity of the radial mobility of ^{137}Cs and ^{90}Sr radionuclides between the rings.

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