DISTRIBUTION OF ¹³⁷CS AND ⁴⁰K IN THE TISSUE OF SILVER FIR TREES (*ABIES ALBA* MILL.) FROM LIKA (CROATIA)

DISTRIBUCIJA ¹³⁷Cs I ⁴⁰K U TKIVIMA STABALA OBIČNE JELE (*Abies alba* Mill.) IZ LIKE (HRVATSKA)

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SUMMARY

The research on activities of ¹³⁷Cs and ⁴⁰K, which was conducted on the silver fir (*Abies alba* Mill.) from Lika has included sampling of the trees in the field (rings of the bole from three different heights separated into bark, growth rings, roots, needles, shoots, and the soil surrounding the cut down trees), laboratory analysis of samples using the gamma-ray spectrometry and the statistical analysis of the collected data. The radial and vertical distribution of cesium (¹³⁷Cs) in trees was investigated. ¹³⁷Cs has contaminated forest ecosystems by remote atmospheric transport and radioactive precipitation as a result of nuclear test including the nuclear accident in Chernobyl. On a longer time scale, the variability of the ¹³⁷Cs distribution determined in the organisms of the silver fir depended on the half-life, while the seasonal dynamics were influenced by the degree of physiological activity and the characteristics and functions of plant tissues. The highest activity of ¹³⁷Cs was determined in the bark and the physiologically most active parts of the silver fir (shoots and needles). The highest activity concentration of the ¹³⁷Cs in the growth rings was measured in the lowest parts of the silver fir trees. This research contributed to understanding the behavior of ¹³⁷Cs, which entered the organisms of dominant tree species in the forest ecosystem, as well as its distribution in time and space.

KEY WORDS: forest ecosystem, radionuclides, distribution, bioindicators, silver fir

INTRODUCTION

UVOD

Forest ecosystems provide the basic requirements for maintenance of all life on Earth. Forests directly involved in the purification of groundwater and surface water, protect reservoirs of drinking water from pollution, play an important role in flood protection, protect the soil from erosion and sliding, and prevent the occurrence of avalanches and sliding snow weight (Prpić et al. 2005). The larger forest areas affecting the climate mitigation of major changes in temperature, increase humidity during summer, prevent changes in the microclimate of a particular area, protect against contaminated air, strong wind and noise, hold large amounts of dust, and provide shelter and food for numerous animal and plant species (EEA 2016). Anthropogenic pollution of forests, forest ecosystems and the overall environment is one of the largest problems of our time, which in many areas takes on catastrophic proportions. Many pollutions in forest ecosystems are monitored systematically, their appearance, performance, duration and disappearance are recorded. The radioactive cesium isotope ¹³⁷Cs appeared in the environment in significant quantities after atmospheric nuclear testing during the 1950's and 1960's. (UNSCEAR 1993, FAS 2002), and a large amount of ¹³⁷Cs

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was discharged into the atmosphere during the nuclear accident at Chernobyl on April 26, 1986.

The deposition of radionuclides on the tree canopies is the most important contaminating form of 137Cs in forest ecosystems (Skoko et al. 2011). Given that the tree canopies are a major part of the biomass in the management of low aerodynamic resistance, they are effective "interceptor" of the radionuclide (Yamagata et al. 1969, Bunzl and Kracke 1988, Desmet and Myttenaere 1988, Sokolov et al. 1990). Generally, such "interception" in the trees (seen in the same unit volume of crown) is more effective for dry than wet deposition, especially for small particles and gases (Pröhl 2008). As the amount of ¹³⁷Cs over time decreases exponentially, adoption root becomes the dominant source of intake of ¹³⁷Cs in the trees long term (Ohashi et al. 2020). Accumulation of ¹³⁷Cs in the trees is one of the most important problems in the wood use and forest pollution (Shinta et al. 2014). Wood as a raw material is permanently present in the environment of people, contributing to their radioactive exposure by increasing doses of radiation. However, compared to the annual equivalent dose per capita from various sources, the calculated annual equivalent doses do not pose a risk in beech, oak and silver fir wood panel samples tested from the territory of the Republic of Croatia (Hus et al. 2004).

The objective of the research was to determine the incidence and intensity of the dynamics of the ¹³⁷Cs in the silver fir tissues (*Abies alba* Mill.), including their by-products entering the human and animal food chain, as well as in samples of soil directly under the selected trees. At study sites, on a longer time scale (2003 - 2017), measured variability of ¹³⁷Cs in the tissue of silver fir trees depend on seasonal dynamics of ¹³⁷Cs followed the level of physiological activity and decay of ¹³⁷Cs during time. The highest activity of ¹³⁷Cs was determined in the bark and the physiologically most active parts of the silver fir (shoots and needles) during the growing season. Activities of ⁴⁰K were of approximately equal values throughout the observed period. This research will contribute to the understanding of the biogeochemical cycle of ¹³⁷Cs within the forest ecosystem, and will also contribute to the monitoring of ¹³⁷Cs wood raw material pollution on wood products. This research also tends to contribute to the study review discussion of the consequences of human activity has on the forest ecosystem.

MATERIAL AND METHODS

MATERIJAL I METODE

Research area – Područje istraživanja

The research was conducted on the tissues of fir trees (*Abies alba* Mill.) in Lika (near Vrhovine), and the soils next to the trees. The chosen location is considered to be among the most contaminated during the Chernobyl disaster in Croatia (Barišić and Lulić 1990), and the sampling schedule is shown in Table 1.

Sample collection method – *Metoda prikupljanja uzoraka*

In the forest ecosystem at the site *GJ Komarnica*, subdivision 12a, studied the trees of silver fir (1, 2, 3) and recorded the coordinates of trees and soils samples. The measured trees were knocked down in the field by licensed workers of the company Hrvatske šume Ltd. after which the cut rings were used for further sectioning. Pits were dug next to fir trees in which root samples were collected for testing. The soil samples were collected along with the tree samples. **The first sampling** on a silver fir tree, 25 m high and 56 cm in (DBH) diameter at breast height, was performed during the vegetation dormancy on December 4, 2003. On a felled

Table 1 Sampling locations with data on sampled trees

 Tablica 1. Lokacije uzorkovanja s podacima o uzorkovanim stablima

Research location <i>Lokacija</i>	Date of sampling <i>Datum uzorkovanja</i>	Tree Stablo	Type of sample <i>Vrsta uzorka</i>
Forest economic unit (GJ) Komarnica, subdivision 12a Gospodarska jedinica (GJ) Komarnica, odsjek 12a	December 4, 2003 4. prosinca 2003.	Silver fir <i>– Obična jela</i> 1 DBH-diameter at breast height <i>– Prsni promjer</i> = 52 cm Tree height <i>– Visina stabla</i> = 25 m Tree age <i>– Starost</i> = 94 γ god.	rings, needles, root and lateral roots, peak shoots, soil along the tree <i>kolutovi,</i> <i>iglice, korijen i postrano korijenje, vršni</i> <i>izbojci, tlo uz stablo</i>
	September 3, 2004 <i>3. rujna 2004.</i>	Silver fir – <i>Obična jela</i> 2 DBH-diameter at breast height – <i>Prsni promjer</i> = 54 cm Tree height – <i>Visina stabla</i> = 24.5 m Tree age – <i>Starost</i> = 101 y god.	rings, needles, root and lateral roots, peak shoots, soil along the tree <i>kolutovi,</i> <i>iglice, korijen i postrano korijenje, vršni</i> <i>izbojci, tlo uz stablo</i>
	March 27, 2017 27. ožujka 2017.	Silver fir – <i>Obična jela</i> 3 DBH-diameter at breast height – <i>Prsni promjer</i> =59 cm Tree height – <i>Visina stabla</i> = 26 m Tree age – <i>Starost</i> = 130 y <i>god.</i>	rings, needles, peek shoots, soil along the tree <i>kolutovi, iglice, vršni izbojci, tlo</i> <i>uz stablo</i>

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tree silver fir rings were taken at a height of 0.1 m, 8 m and 16 m and then stratified on samples of biological material, that is, samples of bark and sapwood, and the growth rings with the time steps to the center of the trunk were set aside. On the sawn-off reel of a felled tree at a height of 0.1 m, rings were singled out in sections of two years for the period 1974 - 2003, in sections of four years for the period 1944 - 1973 and then one sample for the period from 1909 - 1943. The same collection dynamic is used for tree rings samples at altitudes of 8 and 16 meters, provided that at 4 meters the sample covered the period 1958 - 2003, and at a height of 16 meters the period 1978 - 2003. The main root and lateral roots (bark and wood) were sampled, and the group of samples were split according to diameter dimensions (<1 mm, 1-2 mm, 3-4 mm, 5-8 mm, 9-16 mm, 17 - 24 mm and 25 - 38 mm). Twigs (peak shoots) were sampled as individual annual samples for shoots that grew in the period 1994 - 2003 at heights of 8 m and 16 m, and at a height of 25 m in the period 1997 - 2003.

The second sampling on the silver fir tree was done during vegetation period on September 3, 2004. On the felled tree of 24.5 m height and 54 cm breast diameter, the rings were takes as in the first sampling, and then stratified on samples of biological material. On the reel of a felled tree, at a height of 0.1 m, rings were singled out in sections of five years each for the period 1945 - 2004, and in sections of ten years for the period 1903 - 1944. Samples were collected on the rings of a felled tree at heights of 8 and 16 meters at the same dynamics, with the period 1927 - 2004 being covered at eight meters, and the period 1960 - 2004 at a height of 16 meters. The main root and lateral roots (bark and wood), were sampled and separated in groups of samples as well as in the first sampling. Twigs (peak shoots) trees were sampled as individual annual samples for shoots that have grown in the period 1994 - 2004 at the height 8 m and 16 m, and a height of 25 m for the period 1996 - 2004. In the first and second sampling, along with peak shoots, samples of peak shoots needles were collected at the heights of 8 m, 16 m and 25 m.

Third sampling on the tree silver fir was conducted on March 27, 2017 for the purpose of comparing partial results after a long period since the last sampling (12 years). At Komarnica, the forest management unit, in subdivision 12a, felled the tree silver fir 26 m in height and 59 cm breast diameter, and the taken disc was then stratified on samples of biological material, that is, samples of the bark, bark and cambium, and annual rings with time intervals to the center of the trunk were singled out. The samples for this analysis were selected at 0.1 m height as the lowest level of the tree where the active life cycle takes place, and that is comparable to the first two samplings to determine the activity of ¹³⁷Cs. In the period 1974 - 2016, the years were separated into sections of two years each, and for the period 1939 -

1973 in sections of five years, and one sample was separated for the period 1887 - 1938. Twigs (peak shoots) trees were sampled as individual annual samples for shoots that have grown at the height of 26 m in the period 2013 - 2016. Along with peak shoots, samples of peak shoots needles were collected. Composite soil samples were collected with felled trees in 2003, 2004 and 2017 for the analysis of ¹³⁷Cs concentration in soils by gamma spectrometric method. For each analysis, approximately 1 kg of the average sample (taken from multiple points (5), homogenized, disrupted in a state) was collected.

Sample preparation and gamma spectrometric method – *Priprema uzoraka i gamaspektrometrijska metoda*

The activity of all samples was measured by gamma spectrometric method in the Laboratory for Radioecology, Division for Marine and Environmental Research, of the Ruđer Bošković Institute which is accredited for performed gamma spectrometric measurements since 2008, according to HRN EN ISO/IEC 17025. Before the measurements, all samples were homogenized, dried in an oven to constant mass, and then placed in the measuring geometry 125 cm³ volume, weighed and sealed. Canberra's HPGe (High Purity Germanium) detector system was used for measurements, where each sample was measured for 80.000 seconds. The Genie 2000 program from the same manufacturer was used to analyze the recorded gamma spectra. The concentration of ¹³⁷Cs activity in the samples was determined from the photo-peak at an energy of 661.6 keV, and the concentration of 40K activity from the photo-peak at an energy of 1460.7 keV. The reliability of the system was checked during intercomparison measurements regularly.

Since the analyzes showed that the composite uncertainty (U) is predominantly influenced by the uncertainty of the counting rate (U_{bb}), which also includes the uncertainty of the counting rate of the basic radiation, the uncertainty of determining the efficiency (U_{ef}) and the uncertainty of determining the surface of photo-peak (U_{fv} - especially in the case of a small number of registered events) these parameters were taken for the calculation of the measurement uncertainty according to the relation:

$U^2 = U_{bb}^2 + U_{ef}^2 + U_{fv}^2$

Extended measurement uncertainties since 2008 were expressed with an overlap factor k = 2 (95% reliability of the results) as opposed to measurements performed until 2008, when measurement uncertainty calculations included only the uncertainty of determining the photo-peak area with an overlap factor k = 1 (68.27% reliability of the results). The measurement uncertainty was calculated as described above and shown in the results. At lower activities, the measurement uncertainty is high, but we consider the obtained re-

sults to be statistically reliable. The reason for this is that the measurements are made in such a way to ensure the same measurement conditions each time (geometry of the sample, measurement time, the same detector and calibration).

Calculation elimination of radioactive decay impact – Računska eliminacija utjecaja radioaktivnog raspada

Given the large time gap between the first and second sampling and measurements (2003 and 2004) on the one hand, and the third sampling and measurements (2017) on the other hand, it could be assumed that the measured activity of ¹³⁷Cs in the third measurement, due to the half-life of ¹³⁷Cs, would be lower for about a quarter than in the samples from 2003 and 2004. The law of radioactive decay is well known and allows the calculation of radionuclide activity at any point in time. Therefore, it is possible to eliminate the influence by computation of radioactive decay for the measurement of 2017, in a way that all of the measured activity of ¹³⁷Cs in the samples from the three measurements, recalculate for the date of July 1, 2003 allowing the comparison of the first and second measurements, and for July 1, 2016, when approximately one half-life of ¹³⁷Cs has expired since the contamination (1986). All samples were computationally analyzed in the described manner, and the results for all samples were shown and interpreted in total (as an arranged pair of values for July 1, 2003 and July 1, 2016), ensuring that the interpretation of results of the difference between recalculated value of the third sampling and the first and second sampling can be exclusively attributed to geochemical processes that occur in the environment over time (given that the impact of radioactive decay is eliminated by calculation).

RESULTS AND DISCUSSION

REZULTATI I RASPRAVA

Distribution of ¹³⁷Cs and ⁴⁰K in the tissues of silver fir – *Distribucija* ¹³⁷Cs *i* ⁴⁰K *u* tkivima obične jele

Distribution results of ¹³⁷Cs in the tissues of silver fir (*Abies alba* Mill.) refer to the peak shoots and needles, the rings, the bark and cambium and root, and to reviewed comparative presentation of results in all silver fir sampled tissues.

Distribution of ¹³⁷Cs in the peak shoots and needles of silver fir – *Distribucija* ¹³⁷Cs u vršnim izbojcima i iglicama obične jele

Figure 1 shows the distribution of ¹³⁷Cs activity in silver fir trees shoots of different heights and ages in all three sampling years, showing also the distribution according to the age of shoots (and not to the calendar year).

Comparison of age sequences of ¹³⁷Cs activity in shoots of silver fir (Figure 1) for individual height between two years (2003 and 2004), using the non-parametric test for related



Figure 1. Distribution of ¹³⁷Cs in fir shoots for different shoot heights and ages (value 0 for shoot age represents the youngest shoot) for three years of sampling.

Slika 1. Distribucija ¹³⁷Cs u izbojcima obične jele za različite visine i starosti izbojaka (vrijednost 0 za starost izbojka predstavlja najmlađi izbojak) za tri godine uzorkovanja.

samples ("Wilcoxon matched pairs test"), shows that the values of 2004 are statistically significantly (p = 0.05) higher at altitudes of 8 and 16 m than those in 2003, while at the 25 m difference between the two was not statistically significant. This could be interpreted as a result of an increase in the photosynthesis intensity, which is in the fir, as coniferous species, present throughout the year and of the trees metabolism level during the growing period, leading to a temporary accumulation of ¹³⁷Cs in the shoots.

Figure 2 show the distribution of ¹³⁷Cs in silver fir needles at different heights and different age in three sampling years.





Slika 2. Distribucija ¹³⁷Cs u iglicama obične jele za različite visine i starosti iglica (vrijednost 0 za starost odnosi se na iglice s najmlađeg izbojka) za tri godine uzorkovanja.

Comparison of age sequences of ¹³⁷Cs activity in shoots of silver fir with those in silver fir needles (Figure 2) for individual height between two years (2003 and 2004), also using non-parametric test for related samples ("Wilcoxon matched pairs test"), shows that the 2004 values are statistically significantly (p = 0.05) higher than in those in 2003, at all three heights. This result, very similar to that of the shoots, can be equally interpreted (as a result of increasing the rate of photosynthesis and metabolism levels during vegetation period, leading to a temporary accumulation of ¹³⁷Cs in the needles). The regularity is shown even at a height of 25 m (not in the shoot case).

Comparison of age sequences of ¹³⁷Cs activity in silver fir shoots with those in needles (Figures 2 and 3) was conducted separately for each height and year, also using the nonparametric test for related samples. The obtained results show that for the same year the values in the shoots are always statistically significantly (p = 0.05) higher than those in the needles of the same age at all three heights. This result can be associated with a smaller amount of flow in the peripheral tissues of the tree conduction system, which results in smaller amounts of ¹³⁷Cs entering/appearing in these tissues. Figure 3 shows a transparent distribution of ¹³⁷Cs through shoots and silver fir tree needles of different ages, separately for each height (8, 16 and 25 m). This figure shows what has already been pointed out: 1) higher values in 2004 during the vegetation period compared to those in the winter period of 2003 (at all heights, both in shoots and needles) and 2) generally higher values in shoots relative to those in needles (for both years, at all tree heights). Additionally, in this picture (looking only at the medians of age series related to the same tissue type in the same year connected by lines), it can qualitatively determine an overall increase in ¹³⁷Cs activity in shoots and needles of the



Figure 3. A clear comparison of distributions (Box-Whisker diagram) of ¹³⁷Cs values across different ages of fir shoots and needles for three tree heights. The dots show the median values of the age sequences, the rectangles the interquartile range (lower and upper quartiles), while the length represents the range between the minimum and maximum recorded value.

Slika 3. Pregledna usporedba distribucija (Box-Whisker dijagram) vrijednosti ¹³⁷Cs kroz različite starosti izbojaka i iglica obične jele za tri visine stabla. Točke prikazuju medijanu vrijednosti starosnog niza, pravokutnici interkvartilni raspon (donji i gornji kvartil), dok dužina predstavlja raspon između minimalne i maksimalne zabilježene vrijednosti.

tree heights (regardless of age). Although due to the small number of sample point (three heights) it is not possible to test this increase for significantly positive correlation, it is appropriate to point out that such positive correlation is consistent with other results presented. That is, when viewed within the same type of tissue, the largest activity of ¹³⁷Cs can be expected in areas of greatest physiological activities, which, if we talk about the shoots and needles, should be higher in the tissue sample from higher stem (having



Figure 4. Distribution of ¹³⁷Cs activity in all sampled rings of silver fir trees at tree heights of 0.1, 8, and 16 m for the samples from 2003. The abscissa shows the calendar year belonging to a particular year.

Slika 4. Distribucija aktivnosti ¹³⁷Cs u svim uzorkovanim godovima obične jele na visinama stabla od 0,1, 8 i 16 m za uzorke iz 2003. Na apscisi je prikazana kalendarska godina koja pripada određenom godu. less overshadowing impact of their own and other crowns, resulting in more radiated solar energy and higher intensity of photosynthesis).

Distribution of ¹³⁷Cs in the annual rings of silver fir – *Distribucija* ¹³⁷Cs *u godovima obične jele*

Figure 4 shows the distribution of ¹³⁷Cs above the limit of detection in all sampled annual rings of silver fir trees at heights of 0.1, 8 and 16 m for samples from 2003. It is noted that in the annual rings of silver fir at all altitudes present much greater variability of ¹³⁷Cs activity that is measurable way back in the past, reaching to the very center of the three rings (one on each level) from the fir tree felled for this survey in 2003, which was, as well as two trees felled in 2004 and 2017, more than hundred years old.

These long time series of radioactivity values can be divided into four intervals based on a graphic insight into the measured data (Figure 4):

- 1) In the period from the beginning of life trees from 1910 to 1943, constant value of ¹³⁷Cs activity of 28.4 ± 0.9 Bq/kg (both in terms of the radius of the coil and in terms of the height of the tree) was measured.
- 2) The period 1944 to 1958, almost constant (in terms of the radius of the disc and the height of the tree) 137 Cs activity was observed, varying from 22.7 ± 0.8 Bq/kg to 23.3 ± 0.9 Bq/kg,
- 3) For the period 1959 to approximately 1983, variation of 137 Cs activity was also recorded in tree rings (without obvious regularity with regard to the year and the height of sampling), in the interval between 14.3 ± 0.7 Bq/kg, (16 m; 1982) to 36.3 ± 1.2 Bq/kg (8 m; 1974),
- 4) For the period from approximately 1984 to 2003, in which, a continuous increase in ¹³⁷Cs activity was recorded in the youngest rings.

Based on the same results, it could even be assumed that the old xylem tissues that no longer participate in the flow from the root to the canopy represent a kind of accumulation reservoir of ¹³⁷Cs, leaving the question of whether ¹³⁷Cs in these (mostly dead) tissues is permanently excluded from migration within the plant and "captured" (but can disappear only by radioactive decay) or it is a temporary seasonal accumulation during the winter period (in which case, the constancy of the recorded values could explain the assumed constant capacity of dead wood). Additionally, given the existence of zone 3. in the dendrochronological sequence of fir, it can be assumed that the transition between the physiologically active and physiologically inactive part of the xylem in felled fir was very gradual (extending to more than thirty years of the dendrochronological sequence), and moreover, anatomically non-homogeneous (due to the variability of the recorded disc height sampling).

Figure 5 shows the distribution of ¹³⁷Cs activity above the detection limit in all sampled annual rings of silver fir trees at heights of 0.1, 8 and 16 m for samples from 2004.

Graphic insight into the measured data shows the following:

- 1. Measurable ¹³⁷Cs activity is present up to the oldest rings, as with the 2003 annual rings
- 2. The variability of ¹³⁷Cs activity in the dendrochronological sequence from 2004 is lower than in those from 2003, which is mostly due to lower values in the youngest (and physiologically probably the most active) years (2004 compared to 2003), which could be attributed to the fact that 2004 samples were from the vegetation season (as a possible cause of increased migration of ¹³⁷Cs from conductive tissues to the most physiologically active tissues in the canopy) and
- 3. Significantly weaker possibility of defining typical zones within the dendrochronological sequence in relation to the series from 2003, although the zone of increased ¹³⁷Cs values in the youngest rings is also visible here, and, on the other hand, the zone of almost constant ¹³⁷Cs values in the oldest rings is well noticeable (on the basis of which it could be assumed that during the vegetation season there is even a migration of ¹³⁷Cs from the inactive to the physiologically active xylem zone, and in the winter ¹³⁷Cs is temporarily accumulated even in the physiologically inactive part of the xylem zone; compare above for dendrochronological sequence from 2003).

Divided time sequences can be observed at intervals for: a) the period from the beginning of tree life to 1958, in which a similar level (in terms of circle radius and tree height) of 137 Cs activity was recorded, which varied from 15.2 ± 0.8 to 19.4 ± 0.9 Bq/kg at a height of 0.1 m, and from 20.0 ± 1.4 to 23.2 ± 1.5 Bq/kg at a height of 8 m, b) the period of 1959 to approximately 1982, in which a variation in 137 Cs activity was recorded, both in terms of ring radius and in terms of tree height (without distinct regularity with respect to the year formation), ranging between 10.4 ± 0.7 Bq/kg (0.1 m; 1980 - 1984 interval) to 25.1 ± 1.5 Bq/kg (16 m, the interval 1970 - 1974) and c) the period of about 1985 to 2004, in which substantially continuous increase of 137 Cs is recorded in the most recent annual rings.

Figure 6 shows a comparison of ¹³⁷Cs activity in the wood rings of silver fir trees (at a height of 0.1 m) between the average values of samples from 2003 - 2004, on one hand, and recalculated (on July 1, 2003) values from 2017 on the other.

Graphic insight into the measured data shows the following: a) lower values in recalculated samples of 2017 compared to the samples from the period 2003 - 2004, b) still measurable ¹³⁷Cs activity along the dendrochronological sequence from 2017, and c) low variability in ¹³⁷Cs activity



Figure 5. Distribution of ¹³⁷Cs activity in all sampled rings of silver fir trees at tree heights of 0.1, 8 and 16 m for the samples from the year 2004. The abscissa shows the calendar year belonging to a particular year.

Slika 5. Distribucija aktivnosti ¹³⁷Cs u svim uzorkovanim godovima obične jele na visinama stabla od 0,1, 8 i 16 m za uzorke iz 2004. Na apscisi je prikazana kalendarska godina koja pripada određenom godu.



Figure 6. Distribution of ¹³⁷Cs activity in rings of silver fir trees 2003 - 2004: average values from the respective samples collected in years 2003 and 2004; 2017- recalculated: values from the respective samples collected in 2017 recalculated (calculated elimination of the impact of radioactive decay) on July 1, 2003.

Slika 6. Distribucije aktivnosti ¹³⁷Cs u godovima stabala obične jele 2003 - 2004.: srednje vrijednosti iz odnosnih uzoraka prikupljenih 2003. i 2004. godine; 2017.-normirano: vrijednosti iz odnosnih uzoraka prikupljenih 2017. godine normirane (računski eliminiran utjecaja radioaktivnog raspada) na dan 1. 7. 2003.

throughout the 2017 series (including the absence of a more evident increase in the youngest years).

Figure 7 shows the distribution of ¹³⁷Cs in the annual rings of silver fir for different height trees and old growth rings in all three sampling years. These are the same values as in the charts of Figures 4 - 6, additionally displaying the distribution by age rings (and not according to the calendar year), and with a limited set of twenty early years within which the samples from 2003 and the 2004 recorded an increase in ¹³⁷Cs activity with decreasing rings age.

Comparison of age series ¹³⁷Cs activity in the annual rings of silver fir for different heights and year of sampling, was done using non-parametric test for related samples ("Wilcoxon matched pairs test"), Figure 8, and shows the following:

1) At the tree heights of 0.1 m to 8 m statistically significant (p = 0.05) increases were observed in 2003 (while at the height of 16 m this difference was not significant), which can be attributed to a vegetation dormancy in winter and



Figure 7. Distribution of ¹³⁷Cs activity in rings of silver fir for different tree heights and ages (value 0 for age represents the youngest year) for three years of sampling.

Slika 7. Distribucija aktivnosti ¹³⁷Cs u godovima obične jele za različite visine stabla i starosti goda (vrijednost 0 za starost goda predstavlja najmlađi god) za tri godine uzorkovanja.



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Figure 8. A clear comparison of distributions (Box-Whisker diagram) of ¹³⁷Cs values across series of silver fir rings for three tree heights in three sampling years. The dots show the median values of the age series, the rectangles the interquartile range (lower and upper quartiles), while the length represents the range between the minimum and maximum recorded value.

Slika 8. Pregledna usporedba distribucija (Box-Whisker dijagram) vrijednosti ¹³⁷Cs kroz starosne nizove godova obične jele za tri visine stabla u tri godine uzorkovanja. Točke prikazuju medijanu vrijednosti starosnog niza, pravokutnici interkvartilni raspon (donji i gornji kvartil), dok dužina predstavlja raspon između minimalne i maksimalne zabilježene vrijednosti.

2) There was no statistically significant difference between series from the same sampling year, and from different heights, which can be interpreted as a homogeneous height distribution of ¹³⁷Cs activity in the twenty youngest years of the tree rings.

Distribution of ¹³⁷Cs in bark and cambium of silver fir – *Distribucija* ¹³⁷Cs u kori i kambiju obične jele

In Figure 9, (comparing only the same tissue type in the same year) a general increase of ¹³⁷Cs activity in the bark and cambium can be found correlating with the tree height (except at 0.1 m in 2003). In 2003, the highest concentration of ¹³⁷Cs was measured in bark samples collected at a tree height of 16 meters, measuring 130.1 ± 1.5 Bq/kg, while the value of the measured sample at the same height in 2004 was 50.7 \pm 1.2 Bq/kg. At the same height (16 m), the ¹³⁷Cs activity in cambium was 124.1 ± 1.4 Bq/kg measured in 2003, and in 2004 it was 79.2 ± 1.6 Bq/kg. In the samples of the bark on tree height of 8 meters in 2003 measured ¹³⁷Cs activity was 83.8 \pm 1.4 Bq/kg, and in 2004 was 38.7 \pm 1.0 Bq/kg. At the same height (8 m) in 2003 measured ¹³⁷Cs activity in the cambium was 77.6 ± 1.4 Bq/kg, and in 2004 it was 65.2 ± 1.4 Bq/kg. At a height of 0.1 m, the measured ^{137}Cs activity in the bark sample of 2003 was 70.6 \pm 1.3 Bq/ kg, and in 2004 sample it was 24.0 ± 0.3 Bq/kg. At the same height (0.1 m), the ¹³⁷Cs activity in the cambium was 80.5 \pm 1.5 Bq/kg measured in 2003, and in 2004 it was 27.0 \pm 0.3 Bq/kg. The values of the measured ¹³⁷Cs activity in 2017 and recalculated to day July 1, 2003 amounted to 13.8 ± 2.0



Figure 9. Vertical distribution of ¹³⁷Cs in the bark and cambium of silver fir Slika 9. Vertikalna distribucija ¹³⁷Cs u kori i kambiju obične jele

Bq/kg in bark and to 18.4 ± 2.67 Bq/kg, in cambium, recalculated to day July 1, 2016 amounted 10.2 ± 1.48 Bq/kg in bark and 13.6 ± 1.3 Bq/kg in cambium.

Distribution of ¹³⁷Cs in the root of the silver fir – *Distribucija* ¹³⁷Cs u korijenu obične jele

Distribution of ¹³⁷Cs activity in the root thickness classes of silver fir of 2003 and 2004 samples is shown in Figure 10. The samples from the 2003 showed of ¹³⁷Cs activity variation ranging from 62.1 ± 1.3 Bq/kg (root thickness 1-2 mm) to 98.4 ± 1.8 Bg/kg, while in samples from 2004, the range was from 31.4 ± 1.0 Bq/kg (root thickness 3-4 mm) to 71.2 \pm 2.3 Bq/kg (tiniest roots). Meanwhile, the value nearest the maximum (recorded in the thinnest roots) for both years were recorded in the thickest roots (25-38 mm; 95.5 \pm 2.7 Bq/kg in samples from 2003; 44.9 ± 1.5 Bq/kg in samples from 2004) which at the qualitative level indicate the absence of a link between root thickness and ¹³⁷Cs activity. This conclusion is supported by the analysis of the association of ¹³⁷Cs activity with the upper limit of the root thickness class, performed by the Spearman-rank correlation method, suitable for small samples, which did not result in a significant correlation (RSP = 0.1429 with p = 0.7599 for 2003). RSP = - 0.3214 with p = 0.4821 for 2004). Accordingly, it can be concluded that no significant variability in ¹³⁷Cs activity has been observed in the fir root system, which could be due to the fact that these are plant tissues that overgrow the soil from which the tree draws ¹³⁷Cs. By comparing the values of two sampling years separately for each diameter class roots (Figure 10), we can see that in 2003 (winter period) each diameter class recorded higher values than in 2004 (growing season). This result could be explained by the assumption that in winter, because of slower metabolism and less intensive flow through the vascular tissue of roots, roots temporarily accumulate ¹³⁷Cs, whose



Figure 10. Distribution of ¹³⁷Cs activity in thickness classes of silver fir roots (samples from 2003 and 2004)

Slika 10. Distribucija aktivnosti ¹³⁷Cs u debljinskim klasama korijena obične jele (uzorci iz 2003. i 2004. godine)

concentration during the vegetation period decreases due to migration of ¹³⁷Cs in physiologically most active tissues (shoots and needles).

Comparison of ¹³⁷Cs activities in different tissues of silver fir – Usporedba aktivnosti ¹³⁷Cs u različitim tkivima obične jele

Figure 11 shows the distribution of ¹³⁷Cs activity in rings, shoots, needles of silver fir tissues (the results from the youngest tissue samples generated in the sampling year for

tissue types for which is possible to determine the age) for 2003 (winter sample), 2004 (vegetation season sample) and values from the respective samples collected in 2017 and recalculated (eliminated impact of radioactive decay) on July 1, 2003. In Figure 11, it can be primarily observed that in tissues (peak shoots, needles) from samples collected in 2004, values are significantly higher than in previously collected samples, which directly proves the presence of ¹³⁷Cs redistribution in tree tissues over time (given that the absence of such redistribution over time should decrease the value due to radioactive decay). The existence of redistribution is confirmed by the results in the tissues in which the ¹³⁷Cs activity was higher in 2003, because these differences (up to three times higher values for samples of bark and cambium near the soil, compared to 2004) were significantly higher than those that would result due only to radioactive decay during less than a year. Furthermore, it can be seen that in the winter period samples higher values of ¹³⁷Cs were recorded in samples of dead bark, live bark with the cambium, and the young rings and roots, while in the growing season samples higher values were recorded in peak shoots and needles samples. It can be observed that for all tissue types the recalculated values from the samples collected in 2017 are sig-



Figure 11. Distributions of ¹³⁷Cs activity in different tissues of silver fir for 2003 (winter sample), 2004 (vegetation season sample) and 2017- recalculated: values from the respective samples collected in 2017 recalculated (calculated eliminated impact of radioactive decay) on July 1, 2003. For tissue types for which it is possible to determine the age (rings, shoots, needles), only the results from the youngest tissue samples (created in the sampling year) are shown.

Slika 11. Distribucije aktivnosti ¹³⁷Cs u različitim tkivima obične jele za 2003. (uzorak iz zimskog razdoblja), 2004. godinu (uzorak iz vegetacijske sezone) i 2017.-normirano: vrijednosti iz odnosnih uzoraka prikupljenih 2017. godine normirane (računski eliminiran utjecaj radioaktivnog raspada) na dan 1. 7. 2003. Za tipove tkiva kojima je moguće odrediti starost (godovi, izbojci, iglice) prikazani su samo rezultati iz najmlađih uzoraka tkiva (nastalih u godini uzorkovanja)

nificantly less than the relative values from 2003 and 2004, but that these differences are not the same for all tissue types. These results can be explained by tree metabolism slowing down and the intensity of photosynthesis during the vegetation dormancy, which results in poorer flow through the conducting tissues and temporary accumulation of ¹³⁷Cs in the phloem, xylem and root, while on the other hand during the growing season, boosted metabolism and intensive circulation of substances in the relation root - xylem - needles - phloem – root, the temporary accumulation of ¹³⁷Cs is seen in the top shoots and needles.

Relationship of ⁴⁰K and ¹³⁷Cs activities in the tissues of silver fir – Odnos aktivnosti ⁴⁰K i ¹³⁷Cs u tkivima obične jele

Behavior and distribution of cesium in plants and organisms is similar to potassium because these elements are homologous (Shaw and Bell 1991, Robinson and Stone 1992). Research (Lovrenčić et al. 2008) confirms that ¹³⁷Cs is involved in physiological processes in a similar way as potassium. For this purpose, a ⁴⁰K distribution was measured simultaneously with that for ¹³⁷Cs, in the same samples by gamma spectrometric measurement. The scatter plot in Figure 12 shows the ratio of the measured ⁴⁰K and ¹³⁷Cs activities in fir tissues (for twigs, needles, rings, bark and cambium). The obtained result is expressed collectively for all tissue types and shows the ratio of measured values for ⁴⁰K and ¹³⁷Cs, where each point on the graph represents an



Figure 12. Diagram of scattering of measured ⁴⁰K and ¹³⁷Cs activities in silver fir organisms (each point on the graph represents an arranged pair of measured values for ⁴⁰K and ¹³⁷Cs in the same tissue type, sampled in the same year and at the same tree height, if sampled from several heights for a tissue type). The results of correlation analysis (univaried linear regression) were presented within the framework included in the graph.

Slika 12. Dijagram raspršenja izmjerenih aktivnosti ⁴⁰K i ¹³⁷Cs u organizmima obične jele (svaka točka na grafu predstavlja uređeni par izmjerenih vrijednosti za ⁴⁰K i ¹³⁷Cs u istom tipu tkiva, uzorkovanog u iste godine i na istoj visini stabla, ako je za neki tip tkiva uzorkovano s više visina). U okviru uklopljenom u graf doneseni su rezultati korelacijske analize (univarijatna linearna regresija).

Table 2 Activities of ^{137}Cs and ^{40}K (Bq/kg) in composite soil samples at a depth of 0-15 cm

Tablica 2. Aktivnosti $^{\rm 137}Cs$ i $^{\rm 40}K$ (Bq/kg) u kompozitnim uzorcima tala na dubini 0-15 cm

Date of sampling Datum uzorkovanja	⁴⁰K (Bq/kg)	¹³⁷ Cs (Bq/kg)
December 4, 2003 4. prosinca 2003.	$107,0\pm8,4$	274,1 ± 1,7
September 3, 2004 <i>3. rujna 2004.</i>	$144,7\pm8,0$	$\textbf{160,2} \pm \textbf{1,9}$
March 27, 2017 <i>27. ožujka 2017.</i> (on July 1, 2016 <i>na dan 1. 7. 2016</i> .)	$\textbf{240,0} \pm \textbf{31,3}$	57,2 ± 6,08
March 27, 2017 <i>27. ožujka 2017.</i> (on July 1, 2003 <i>na dan 1. 7. 2003.</i>)	240,0 ± 31,3	77,2 ± 8,21

arranged pair of values for ⁴⁰K and ¹³⁷Cs measured in the same tissue type, sampled at the same time (the same year) and at the same height of an ordinary fir tree (when it was sampled from several heights for some type of tissue). We can observe a large scattering of data, with a linear correlation that is statistically significant (R = 0.4999; with p = 0.000), so this result is not inconsistent with the fact that ⁴⁰K and ¹³⁷Cs are homologous isotopes that the plant does not distinguish. The grouping of points from samples of different tissue types can also be observed on the same graph, so it was appropriate to perform the same analysis separately for individual tissue types.

Distribution of ⁴⁰K and ¹³⁷Cs in the soil – *Distribucija* ⁴⁰K i ¹³⁷Cs u tlu

While the focus of the research was the distribution of ¹³⁷Cs activity in the plant tissue of silver fir, we collected composite samples of forest soil at the same site Vrhovine, on which we measured activity of ⁴⁰K and ¹³⁷Cs (Bq/kg) by gammaspectrometer in all three sampling years (Table 2). The level of ¹³⁷Cs activity in the fir forest was much higher in the samples of soil collected in 2003 and 2004 in relation to the activity levels measured on 2017 samples (recalculated on day July 1, 2003 and July 1, 2016). The ⁴⁰K activity level in 2017 was twice higher as in 2003 and 2004.

Activity of ¹³⁷Cs in the tissues of silver fir trees – Aktivnost ¹³⁷Cs u tkivima drveća obične jele

The results of this study showed that the distribution of ¹³⁷Cs activity within each individual tissue was causally related to the degree of physiological activity in a particular tissue. In the radial distribution, this is reflected in an increased activity value with decreasing age (the maximum activity was observed in the youngest wood rings), while the vertical distribution is reflected in the increased activity correlated to the height of the tree. In the underground part (root samples) the similar dependence was measured in the vegetation period, while during the vegetation dormancy the level of ¹³⁷Cs activity did not vary significantly with the reduction of the root dimensions. The results of this study show similar dynamics of ¹³⁷Cs movement in fir tissues (*Abies Alba Mill.*) as in previous results by Popijač et al. (2004) fifteen years

after the Chernobyl accident on Medvednica (Sljeme) and Lovrenčić et al. (2008) in Gorski Kotar in which they monitored the dynamics of ¹³⁷Cs movement on a monthly basis throughout the year and found higher ¹³⁷Cs activities in the bark of fir and heartwood during the growing season, when the trees were physiologically more active. The previous results of the two studies suggest that silver fir is a good model species for ¹³⁷Cs biomonitoring because they behave similarly in different habitats, although this claim would need to be extended to more localities.

Relationship between ⁴⁰K and ¹³⁷Cs activities in trees and soil – Odnos aktivnosti ⁴⁰K i ¹³⁷Cs u drveću i tlu

To understand the possible explanations of long-term presence of ¹³⁷Cs in the forest ecosystem, this study, measuring concentration levels of ¹³⁷Cs activity in the tissues of silver fir and soils near sampled trees, also covered the level of ⁴⁰K activity, trying to explain the relationship between ⁴⁰K and ¹³⁷Cs activities in trees and soil, considering that the two isotopes are homologous. Research (Lovrenčić et al. 2008) also confirms that ¹³⁷Cs is involved in physiological processes in a similar way as potassium, since these two elements are homologous. Their activity is inversely proportional to the age of the needles and twigs, that is, the highest is in the youngest tissue sections. The soil characteristics play a key role in the transfer of ¹³⁷Cs, based on their texture, ability to exchange cations and organic matter content. Clay soils accumulate ¹³⁷Cs and it was found that if the proportion of clay in the soil is higher, the ¹³⁷Cs activity in the soil is also increased, because its sorption is conditioned by clay minerals (Kruyts and Delvaux 2002, Stauton et al. 2002). The content of K⁺ in the soil can cause the collapse of the extended interlayers (Rigol et al. 2002), because in this case the ¹³⁷Cs is binding within the interlayer and is blocked, and unavailable for transfer processes. Potassium availability is strongly associated with sorption and desorption processes, as well as fixations that take place in the soil (Đurđević 2014). Previous research on the relationship of ⁴⁰K and ¹³⁷Cs in soils has been primarily related to measuring and determining the distribution and level of ¹³⁷Cs activity in soils after the Chernobyl and Fukushima accidents (Smolders et al. 1997, Rochon et al. 1998, Gerzabek 1996, Zhu and Smolders 2000, Kruyts and Delvaux 2002; Kaunisto et al. 2002; Zibold et al. 2009). Their importance was highly related to dealing with the consequences that occurred after the entry of ¹³⁷Cs in the environment and taking measures to secure food production and animal farming, but also protect forest ecosystems.

CONCLUSIONS

ZAKLJUČCI

The study of distribution of ¹³⁷Cs in the trees of silver fir (*Abies alba* Mill.) from Lika is based on three occasions within the interval 2003 - 2017 which included field ring

samples of trees with three heights (separated on the bark and growth rings), roots, needles, peaks of shoots, and soils near felled trees, and after laboratory processing of collected samples in the gamma spectrometer, statistical analysis and interpretation of the data obtained, resulted in the following conclusions:

1. Following the results of previous researches in a similar but wider area (which was limited to ¹³⁷Cs activity in soil and honey, Barišić et al. 2018), this study confirmed longterm contamination of Lika forest ecosystems with radioactive precipitation that had entered the Vrhovine area by remote atmospheric transport after the nuclear tests and the Chernobyl nuclear power plant accident, which manifested itself in measurable ¹³⁷Cs concentrations in plant tissues at least three decades after contamination.

2. Throughout the observed period (2003 - 2017), the variability of 137 Cs distribution in the tissues of silver fir trees decreased continuously. The reason for this was on one hand radioactive decay (about 26% in all tissues during the observed period), while on the other hand it was due to the gradual elimination of 137 Cs from tree tissue, mostly through dead bark, and needles, which caused a decrease in 137 Cs activity during the observed period up to 96% in needles, peak shoots and fir rings (in relation to the recalculated values, after elimination of the influence of radioactive decay).

3. The highest ¹³⁷Cs activity of silver fir tissues were measured in peak shoots and needles, and significantly lower activity in other tissues, regardless of the time of year. The conclusion is that the main elimination pathways of ¹³⁷Cs are needles typically several years old and which the tree gradually sheds throughout the year.

4. In the soil habitat of the silver fir ¹³⁷Cs migrated deeper, towards the root, and thus become biologically available.

5. The recorded concentration of ¹³⁷Cs and ⁴⁰K are statistically significant correlations for the total sample, and separately for the needles sample. These results (given that no statistically significant negative correlation was observed, while a significant positive correlation can still be interpreted by a random process) suggest that the tree does not distinguish between these two homologous elements.

6. The results of the research (the first of its kind in Croatia focused on the edificatory tree species tissue) contribute to the understanding of future of ¹³⁷Cs that entered the tissue of edificatory tree species in the forest ecosystem, as well as its distribution in time and space, which (especially in comparison with rare similar research in the world) complements the emergence of the ¹³⁷Cs biogeochemical cycle in the environment (especially in forest trees).

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SAŽETAK

Istraživanje aktivnosti ¹³⁷Cs i ⁴⁰K provedeno na stablima obične jele (Abies alba Mll.) iz Like uključilo je terensko prikupljanje uzoraka (kolutove stabala sa tri visine i razdvajanih na koru i godove, korijen, iglice, vršne izbojke i tla uz oborena stabla), laboratorijsko mjerenje aktivnosti ¹³⁷Cs i ⁴⁰K u prikupljenim uzorcima gamaspektrometrijskom metodom i statističku obradu dobivenih podataka. Ispitivane su radijalne i vertikalne raspodjele cezija (¹³⁷Cs) u stablima koji je kontaminirao šumske ekosustave putem daljinskog atmosferskog transporta i oborina kao posljedice nuklearnih pokusa, kao i havarije u Černobilu. Na dužoj vremenskoj skali utvrđena je varijabilnost distribucije ¹³⁷Cs u tkivima stabala obične jele koja je u ispitivanom razdoblju ovisila i o vremenu poluraspada, dok je sezonska dinamika zavisila o razini fiziološke aktivnosti, te od karakteristika i funkcija biljnih tkiva. Najveća koncentracija aktivnosti ¹³⁷Cs u tvrđena je u kori i fiziološki najaktivnijim dijelovima stabala obične jele (vršnim izbojcima i iglicama). Najveća aktivnost ¹³⁷Cs u godovima obične jele izmjerena je na najnižoj visini stabala. Ovo istraživanje doprinosi razumijevanju ponašanja ¹³⁷Cs koji je ušao u organizam edifikatorske vrste drveća u šumskom ekosustavu, kao i njegove distribucije u vremenu i prostoru.

KLJUČNE RIJEČI: šumski ekosustav, radionuklidi, distribucija, bioindikatori, obična jela