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Original scientific paper

Consequences for the Industry of Radioactive Contamination Accompanied by the Dynamics of ^{137}Cs and ^{40}K Movement in the Environment

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Abstract: Radioactive pollution entered the ecosystems during the nuclear test in the 1950s and 1960s and in Croatia largely after Chernobyl accident in 1986. ^{137}Cs and ^{40}K can be stored in different parts of the trees but also in the upper surface layer of soil under the trees. This study was aimed at determining the intensity and dynamics of ^{137}Cs and ^{40}K radionuclides in chestnut tree tissues (*Castanea sativa* Mill.) in the vicinity of Petrinja (Croatia). The samples were taken in 2003, 2004 and 2016 and the samples activities were measured by gamma spectrometric method. During the winter, the highest activities were recorded in the top shoots, leaves, fruits and hedgehogs, while in the vegetation season, the highest activities were in the top shoots and dead bark. Also, a decrease in ^{137}Cs concentrations was observed in the rings with increasing age of the rings. Significant increased values of activity were measured in samples of the youngest and physiologically most active parts of trees, compared to the least physiologically active where concentrations were lower. ^{137}Cs was biogeochemically retained almost entirely in the surface layer. The obtained results suggest that the chestnut tree does not distinguish ^{137}Cs and ^{40}K, as two homologous elements.

Keywords: ^{137}Cs; ^{40}K; chestnut; distribution; dynamics; environment; radioactive pollution

1 INTRODUCTION

Pollution with the radioactive isotope ^{137}Cs appeared as a result of atmospheric nuclear tests in the 1950s and 1960s [1, 2], and large amounts of ^{137}Cs were released into the atmosphere during the Chernobyl accident, after which radioactive particles reached even Croatia [3]. ^{137}Cs remained stored by dry and wet deposition [4], and ^{137}Cs entered forest ecosystems foliarly and through the root system [5]. The main part of the dry deposited radionuclides was retained in the tree canopy [6], incorporated into the leaf surfaces and moved to other structural components of the tree. By precipitation leaching from canopies, by falling of branches, needles or leaves, ^{137}Cs is also stored in the upper surface layer of soil under trees [6, 7], which retain ^{137}Cs for a long time and are the main source of radioactive contamination of forest vegetation [8, 9]. It can be said that forest ecosystems are closed systems for ^{137}Cs that can disappear only by radioactive decay ($T_{1/2}$ = 30.17 years), physical removal due to human exploitation of forest products (wood, fruit, mushrooms, wild animals) and soil erosion [8, 10].

After the deposition of ^{137}Cs on the organic layer of the soil and the decomposition of organic material by organisms (animals, bacteria, fungi), migration occurs to deeper layers from where ^{137}Cs can be absorbed by the roots and returned to living parts of tree organisms [8]. In this way the cycle of circulation is closed [11]. Bioavailability of ^{137}Cs (expressed by a bioavailability factor that considers the vertical distribution in the soil and the root biomass distribution in its different horizons) is the basis for the analyses of ^{137}Cs transfer from soil to trees in different tree species and in different forest ecosystems [7].

The artificial radionuclide ^{137}Cs is a member of the same homologous series to which the natural radionuclide ^{40}K belongs and, although ^{40}K is a natural radionuclide, the competitive effects of caesium and potassium cannot be ruled out [12]. This assumption is confirmed by research [13]: ^{137}Cs is involved in different physiological processes in a way similar to ^{40}K. This is somewhat expected since caesium and potassium are homologous elements.

The large heterogeneity of ^{137}Cs concentrations in different parts of trees is due to diverse tree tissue metabolisms, and the uneven distribution of ^{137}Cs along the trunk can be explained by fixation of radionuclides on xylem vessel walls and changes in trunk geometry [14]. It was found that ^{137}Cs radial distribution in the trunk depends on ^{137}Cs availability in the soil, which controls the transfer of radionuclides by xylem as well as by the properties of xylem. So, the accumulation of ^{137}Cs in trees is influenced by ^{137}Cs vertical distribution and availability in soil [14, 15]. Treated wood, as a raw material, is constantly present in the human environment, contributing to its radioactive contamination with increased radiation doses. However, if it is compared to the annual equivalent dose per capita (from different sources), it seems that the calculated annual equivalent doses do not pose a risk in wood panels tested by beech, oak and fir samples from the Croatian territory [16].

The aim of this study is to determine the intensity and dynamics of ^{137}Cs and ^{40}K radionuclides in chestnut tree tissues (*Castanea sativa* Mill.) as wood raw materials and the impact on wood products (including by-products) that enter the human and animal food webs, as well as soil samples immediately adjacent to selected trees, all for the purpose of better understanding that part of the ^{137}Cs and ^{40}K biogeochemical cycle that takes place in the forest ecosystem.

2 MATHERIAL AND METODS

2.1 Research Area

The research was conducted in the forest ecosystem of chestnut coppices in the vicinity of Petrinja in Banovina (Tab. 1). The investigated site is considered to be one of the most heavily contaminated area in Croatia after the Chernobyl accident [17]. Samples were collected from the same individual because three shoots grew from the same coppice and developed into the sampled three trunks. On felled trunks, samples of biological material were taken to measure ^{137}Cs and ^{40}K activity in plant tissue and soil samples at the same location.

Table 1 Sampling locations (with sampled trees data)

Sampling location: Economic unit Vučjak - Tješnjak, section 47a		
Sampling date	Samples of chestnut trees	Type of sample
2/12/2003	Chestnut tree 1 Chest diameter = 18 cm; Tree height = 12 m; Age = 23 years	rings, leaves, apical shoots, soil next to the tree
23/6/2004	Chestnut tree 2 Chest diameter = 19 cm; Tree height = 14 m; Age = 55 years	
15/10/2016	Chestnut tree 3 Chest diameter = 21 cm; Tree height = 16 m; Age = 67 years	

2.2 Sampling, Preparation, Measurement and Analysis

The first sampling on the chestnut tree 1 was performed during the dormancy of the vegetation, and the second sampling on the tree 2 during the vegetation. On both occasions, the reels were taken and then stratified into biological material samples, i.e. individual samples from dead and live bark, cambium, and rings from the reels were separated in time intervals of one year for the first three youngest rings and for two years to the centre of the trunk at heights of 0.2 m, 4 m and 8 m. The third process of taking the samples on tree 3 was carried out to compare part of the results after a longer period from the last sampling (more than 12 years), also at the same location, on a tree that grew from the same coppice as for the two previous samples. The samples were analysed in January 2017. A reel at a height of 4.0 m was taken from the felled tree and then stratified into biological material samples. Samples of dead and live bark, cambium and rings with the same time intervals as in the first two sampling were isolated.

2.3 Gamma Spectrometric Analysis

For gamma spectrometric analysis of ^{137}Cs and ^{40}K concentrations in soil, composite soil samples were collected both in 2003 and 2017, and soil samples for determination of the percentage of clay, the amount of organic matter in the soil and potassium concentrations were collected in 2017 near the felled trees. For the needs of the analyses, approximately 1 kg of an averaged sample (taken from five places, homogenized, in a disturbed condition) was collected for each individual analysis.

The activities of all samples were measured by gamma spectrometric method at Ruđer Bošković Institute, Laboratory of Radioecology of the Division for Marine and Environmental Research (Zagreb). Before measurement, each sample was homogenized, dried in a dryer to constant weight, and then placed in vessels with a volume of 125 cm^3, weighed and hermetically sealed. The measurements were performed with Canberra HPGe detecting system and each sample was measured for 80,000 seconds. The analyses of obtained gamma spectra was performed with Genie 2000 programme (from the same producer). ^{137}Cs activities in the samples were measured from 661.6 keV photo-peak while ^{40}K activities were measured from 1460.7 keV photo-peak. The reliability of measurements was regularly checked during the determinations.

Calculation of the measurement uncertainty (U) according to the relation (1):

$$U^2 = U_{bb}^2 + U_{ef}^2 + U_{fv}^2 \quad (1)$$

includes countdown rate uncertainty (U_{bb}), efficiency determination uncertainty (U_{ef}) and photo-peak area determination uncertainty (U_{fv}) especially for small number of registered events and is shown in Results and Discussion.

Since 2008 the extended measurement uncertainties are expressed by the overlap factor k = 2 (meaning 95% reliability of the results). Until 2008 the measurement uncertainty calculations included only photo-peak area determination uncertainty with an overlap factor k = 1 (meaning 68.27% reliability of the results). Although at lower radioactive activities the measurement uncertainty is high, the obtained results are considered statistically reliable because the measurements are performed in such a way that ensures the same conditions each time (measurement time, sample geometry, same detector and calibration).

2.4 Computational Zeroing of the Radioactive Decay Effects

With considering the large time interval between the first and second sampling and measurement (in 2003 and 2004) on one side and the third sampling and measurement (2016/17) on the other side, it could be supposed in advance that the measured ^{137}Cs activities from the third measurement would be approximately a quarter lower than in the 2003 and 2004 samples, due to the half-life decay of radionuclides. For measurements from 2016/17, it is therefore possible to calculate the impact of radioactive decay by calculating all measured mass activities of ^{137}Cs in the samples from the third measurement to date of July, 1 2003 for mutual comparison with the first and second measurements, and also to July 1, 2016, when approximately one half-life of ^{137}Cs had elapsed since the original contamination (in 1986).

In the described way all samples were numerically analysed and the obtained results for all samples were interpreted and presented collectively (as an ordered pair of values for July 1, 2003 and July 1, 2016). The above mentioned ensured that in the interpretation of obtained results the differences between the re-calculated values of the last sampling and the first and second sampling can be attributed entirely to the action of geochemical processes that take place in the forest environment over time (with the impact of radioactive decay eliminated by numerical calculations).

2.5 Analysis of Clay Content, Amount of Organic Matter and Potassium Concentration in Soil

Determination of the percentage of clay, the amount of organic matter in the soil and the concentration of potassium was performed in the laboratory of the Faculty of Agrobiotecnical Sciences in Osijek, since it was expected that these parameters very significantly affect the activity of ^{137}Cs and ^{40}K in chestnut tissues.

3 RESULTS AND DISSCUSION

3.1 Distribution of ^{137}Cs and ^{40}K in Chestnut Tree Canopy Tissues

The ^{137}Cs activities measured in canopy tissues (Tab. 2) were the highest in samples of chestnut hedgehogs collected in 2003 (14.0 ± 0.6 Bq kg^{-1}). In that year, the activities of ^{137}Cs in other tissue types were relatively close to the values measured in hedgehogs, with the highest being in peak shoots (12.1 ± 0.5 Bq kg^{-1}), slightly lower in fruits (10.6 ± 0.4 Bq kg^{-1}), and the lowest in leaves (10.0 ± 0.5 Bq kg^{-1}). In the samples from 2004, the measured values of ^{137}Cs were significantly lower than in the previous year in all tissue types (in hedgehogs 3.6 ± 0.4 Bq kg^{-1}, in fruits 6.0 ± 0.3 Bq kg^{-1}, and in leaves 3.4 ± 0.3 Bq kg^{-1}), except for peak shoots where ^{137}Cs activity of 13.2 ± 0.6 Bq kg^{-1} was measured that year. It should be taken into account that the samples in 2003 were collected in the winter period of dormancy of vegetation with reduced physiological activity in all types of tissues, especially in those that the tree had just rejected (fruits, hedgehogs, leaves), while the samples from 2004 were collected in the vegetation period marked by the highest physiological activity in all these tissue types. For this reason and only on the basis of these results the existence can be assumed of intensive migration of ^{137}Cs to those tissues where the level of physiological activity is maximized at some point. In the same way goes the interpretation of the fact that in 2004 in the tissue of the fruit was recorded almost twice the concentration of ^{137}Cs than in hedgehogs, given that at the time of sample collection the fruit was in the phase of intensive maturation. In 2003, it was the other way around, i.e. the level of ^{137}Cs activity in hedgehogs, whose development ended after the fruit matured, was higher than in the fruit. The recorded activity of ^{137}Cs in peak shoots grown in 2016, normalized on July 1, 2003, was 2.65 ± 1.08 Bq kg^{-1}, which was only 21% of the value recorded at the time of the first two sampling (2003 and 2004), suggesting that nearly four-fifths of ^{137}Cs were lost from this tissue type by biogeochemical processes during the observed period. On the other hand, the recorded (and equally normalized) activity in leaves grown in 2016 (3.8 ± 1.2 Bq kg^{-1}) was 56% of the value recorded in the first two samplings, which suggests that the activity of ^{137}Cs over time under the influence of biogeochemical processes decreases much more slowly in the leaves than in the shoots. This is also a likely consequence of greater physiological activity in the leaves (through which, by constant migration into this tissue type from season to season, the concentration of ^{137}Cs in other tissue types gradually decreases). In 2003, ^{40}K activities in chestnut fruits of 202.4 ± 9.2 Bq kg^{-1}, slightly lower in hedgehogs 163.6 ± 9.3 Bq kg^{-1}, and approximate values in leaves were measured in canopy tissues 108.6 ± 8.2 Bq kg^{-1} and peak shoots 102.7 ± 7.8 Bq kg^{-1} (Fig. 1). In 2004, the measured values of ^{40}K in peak shoots were 252.6 ± 10.3 Bq kg^{-1}, in leaves 246.4 ± 10.5 Bq kg^{-1}, in hedgehogs 155.3 ± 9.1 Bq kg^{-1} and in fruits 148,0 ± 7.4 Bq kg^{-1}. Recorded ^{40}K activity in peak shoots grown in 2016 was standardized on 1 July 2003 in the amount of 165.0 ± 29.8 Bq kg^{-1}, and in leaves 192.0 ± 33.6 Bq kg^{-1}.

Table 2 Activities of ^{137}Cs and ^{40}K (Bq kg^{-1}) in branches, fruits, leaves and hedgehogs of chestnut

Sample	^{40}K (Bq/kg)	^{137}Cs (Bq/kg)
2003		
The tops of the branches grew in 2003	102.7 ± 7.8	12.1 ± 0.5
Chestnuts fruits in 2003	202.4 ± 9.2	10.6 ± 0.4
The leaves fell off in 2003	108.6 ± 8.2	10.0 ± 0.5
Hedgehogs of fruits in 2003	163.6± 9.3	14.0 ± 0.6
2004		
The tops of the branches grew in 2004	252.6 ± 10.3	13.2 ± 0.6
The tops of the branches grew in 2003	132.0 ± 8.0	9.1 ± 0.5
The tops of the branches grew in 2002	128.8 ± 8.2	7.2 ± 0.5
Chestnuts fruits in 2004	148.0 ± 7.4	6.0 ± 0.3
The leaves fell off in 2004	246.4 ± 10.5	3.4 ± 0.3
Hedgehogs of fruits in 2004	155.3 ± 9.1	3.6 ± 0.4
2016		
The tops of the branches grew in 2016 ^{137}Cs and ^{40}K (on 1/7/2003)	165.0 ± 29.8	2.65 ± 1.08
The leaves fell off in 2016 ^{137}Cs and ^{40}K (on 1/7/2003)	192.0 ± 33.6	3.78 ± 1.21
The tops of the branches grew in 2016 ^{137}Cs and ^{40}K (on 1/7/2016)	165.0 ± 29.8	1.96 ± 0.80
The leaves fell off in 2016 ^{137}Cs and ^{40}K (on 1/7/2016)	192.0 ± 33.6	2.80 ± 0.90

3.2 Distribution of ^{137}Cs and ^{40}K in Chestnut Rings

The activity of ^{137}Cs in the samples of chestnut rings was above the limit of detection only in the first few youngest rings which are also the most physiologically active parts of the tree xylem. This was the case in all three samplings (2003, 2004 and 2016). The highest levels of ^{137}Cs activity in the rings measured in 2003 were in the youngest rings at a height of 4 meters (2.8 ± 0.6 Bq kg^{-1}), slightly lower levels at a height of 8 meters (2.7 ± 0.3 Bq kg^{-1}), and the lowest levels in rings at the ground were at a height of 0.2 meters (1.9 ± 0.4 Bq kg^{-1}). In the measurements carried out in 2004, the highest levels of ^{137}Cs activity in the youngest rings were at the ground at a height of 0.2 meters (2.4 ± 0.9 Bq kg^{-1}), lower levels at a height of 8 meters (1.1 ± 0.5 Bq kg^{-1}), and the lowest levels in years at a height of 4 meters (0.8 ± 0.4 Bq kg^{-1}). Measurements in the same year (2004) but in the rings from the previous year showed a different level of distribution of ^{137}Cs activity at heights of 4 and 8 meters, and the level of ^{137}Cs activity in the previous year (2003) at a height of 4 m was 1.7 ± 0.4 Bq kg^{-1}, and at a height of 8 m was 3.2 ± 0.6 Bq kg^{-1}. ^{137}Cs activity levels in chestnut rings measured in samples collected in 2016 were above the detection limit (^{137}Cs activity level measurements greater than 0.3 Bq kg^{-1}) only for the two youngest rings, with higher value in the youngest ring (0.93 ± 0.70 Bq kg^{-1} on July 1, 2016), or 1.26 ± 0.94 Bq kg^{-1} for the hypothetical value calculated on July, 1 2003 (elimination of the impact of radioactive decay).

The ^{40}K activity in chestnut rings samples was constantly above the detection limit in all ring's samples. The measured values of ^{40}K in external rings ranged from 54.7 ± 9.1 Bq kg^{-1} to 85.8 ± 19.0 Bq kg^{-1}, and in the centre of the trunk from 25.1 ±11.8 Bq kg^{-1} to 35.1 ±13.9 Bq kg^{-1}.

3.3 Distribution of ^{137}Cs and ^{40}K in Chestnut Bark and Cambium

The activity levels of ^{137}Cs and ^{40}K in live and dead bark and chestnut cambium are shown in Fig. 1. Comparing the values of ^{137}Cs in dead bark along the chestnut tree, it can be seen that in 2003 the highest activity was measured at a height of 0.2 meters (8.0 ± 0.5 Bq kg^{-1}), then slightly lower at a height of 8 m (6.8 ± 0.7 Bq kg^{-1}), and the lowest at a height of 4 m (5.8 ± 0.4 Bq kg^{-1}). The highest ^{40}K activity in the dead bark was in the sample since 2003 at a height of 8 m with a measured value of 90.4 ± 11.3 Bq kg^{-1}, and the lowest of 29.7 ± 6.1 Bq kg^{-1}. ^{137}Cs activities in the live bark measured in 2003 had a different vertical distribution from the dead bark of the same year. The highest activity was measured at the highest part of the trunk at a height of 8 m (6.5 ± 0.5 Bq kg^{-1}), and at a height of 0.2 m (6.1 ± 0.5 Bq kg^{-1}) and 4 m (6.1 ± 0.6 Bq kg^{-1}) the same levels of ^{137}Cs activities in the live bark were measured. In live bark samples, the highest value of ^{40}K was measured in the sample at a height of 8 m in 2003, and the lowest in 2004, at a trunk height of 8 m of 55.3 ± 5.5 Bq kg^{-1}. In the vertical distribution of ^{137}Cs activity in the dead bark measured in 2004, the values were higher, and at a height of 0.2 m the ^{137}Cs activity was measured at 22.1 ± 0.8 Bq kg^{-1}, at a height of 4 m the activity of 12,4 ± 0.5 Bq kg^{-1}, and at a height of 8 m activity of 4.2 ± 0.4 Bq kg^{-1}.

The ^{137}Cs activities in the live bark measured in 2004 had the same vertical distribution as in the dead bark of the same year. The highest activity was measured at the lowest part of the trunk, and the lowest at the highest part, at a height of 8 m.

The highest activity of ^{137}Cs in the cambium along the chestnut tree in 2003 was measured at a height of 0.2 meters (3.8 ± 0.4 Bq kg^{-1}), at a height of 4 meters it was 3.3 ± 0.4 Bq kg^{-1}, while at a height of 8 meters it was 4.0 ± 0.5 Bq kg^{-1}. In 2004, there was a significant decrease in height (at 0.2 m activity of ^{137}Cs of 4.1 ± 0.8 Bq kg^{-1}, at a height of 4 meters 1.9 ± 0.8, and at a height of 8 m ^{137}Cs activity of 0.9 ± 0.4 Bq kg^{-1}). The highest activity of ^{40}K (70.9 ± 15.7 Bq kg^{-1}) in the cambium along the chestnut tree in 2003 was measured at a height of 8 m, and the lowest (50.2 ± 12.3 Bq kg^{-1}) of the same year at a height of 0.2 m, while in 2004 the highest activity ^{40}K (113.6 ± 14.4 Bq kg^{-1}) was measured at a height of 0.2 m and the lowest in 2016 (< 35.8 Bq kg^{-1}).

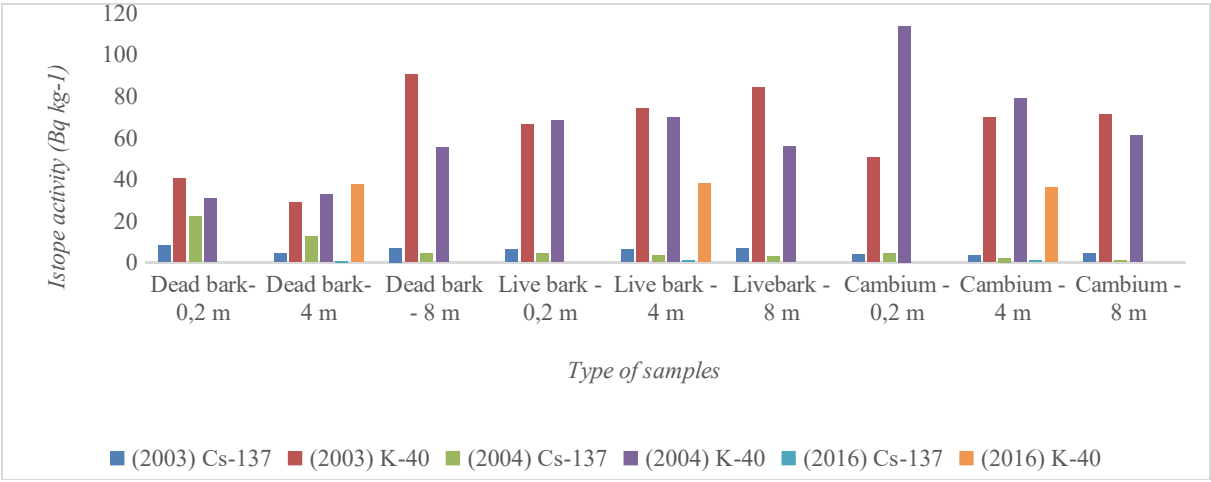

Figure 1 ^{137}Cs and ^{40}K activities measured in chestnut bark and cambium

Regarding the comparison of ^{137}Cs activity values in different tissues of the conduction system, although statistical testing of differences with respect to a small sample is not possible here either, it is possible to perform a qualitative interpretation of the results. It can be observed in Fig. 1 that in both years the values recorded in the cambium were lower than those in the dead bark (which is the case at each individual height) and lower or equal to those in the live bark. At the same time, ^{137}Cs activity levels measured in cambium were an order of magnitude lower than ^{137}Cs activity levels in dead bark, but still higher than in rings, suggesting that cambium, like xylem, takes ^{137}Cs primarily from soil solution through conductive tree system upward flow from root to canopy. An additional argument for such an assumption is the fact that in 2003 (vegetation dormancy) the ^{137}Cs values in the cambium were equal at all tree heights, which can be attributed to reduced physiological activity throughout the tree, while in 2004 (vegetation season) these values were lower as the height of the tree increases, which (similar to the youngest rings) can be attributed to the gradual accumulation of new amounts of ^{137}Cs from the soil (more as the part of the cambium tissue is closer to the ground).

Measured activities of ^{137}Cs in samples of dead and live bark from 2016, standardized on July 1, 2003, in the amount of 0.73 ± 0.62 Bq kg^{-1} for dead, and 0.84 ± 0.45 Bq kg^{-1} for live bark were at the level of only 8% (dead bark) and 18% (live bark) activities recorded in sampling of the same tissue types in 2003 and 2004. In contrast, the normalized activity in the 2016 for cambium was 1.19 ± 0.85 Bq kg^{-1}, reaching almost 46% of that recorded in the same tissue type in the 2003 and 2004 samples. Such a large difference further supports the assumption of a constant supply of cambium tissue with new amounts of ^{137}Cs from the soil. In addition, this difference can be related to the retention of ^{137}Cs during the growing season in the leaves (which the tree will discard at the end of the season, thus evacuating the accumulated ^{137}Cs from the tissue), which also means reducing the concentration of ^{137}Cs in the downstream through phloem. Finally, this difference can probably be associated with the constant loss of ^{137}Cs from the organism by the death and peeling of

the bark, which can be attributed to the relatively rapid elimination of ^{137}Cs from plant tissues.

Regarding the relationship between the recorded ^{137}Cs activities in the live and dead bark, there is a significant difference between the samples from 2003 and those from 2004. Namely, while in 2003 the values in live and dead bark were approximately the same at heights of 4 and 8 m, i.e. approximately 31% more in dead bark at a height of 0.2 m, in 2004 the values in dead bark were many times higher surpassing those in the live bark at heights of 0.2 m (approximately 5.4 times) and 4 m (approximately 3.8 times), while at a height of 8 m they were approximately 56% higher (with a significant increase in values by decreasing height). It can be assumed that such results can also be attributed to differences between samples from the vegetation season (while there was an intensive circulation of substances by way of root - xylem - leaf mass - phloem - root) and samples from the dormant period of vegetation (with temporary interruption of this circulation due to absence of leaf mass). This interpretation would lead to the conclusion that in the part of the ^{137}Cs biogeochemical cycle in the environment that takes place in trees like chestnut, dead bark is a kind of ^{137}Cs accumulator (with a continuous decrease in accumulated amounts over time due to its peeling). It remains unclear whether (and to what extent) this accumulation is temporary (seasonal) with the possibility of re-taking ^{137}Cs by the conducting system of the tree (e.g. in conditions of reduced flow through the phloem), or it is a permanent exclusion of ^{137}Cs from circulating through the plant and its accumulation in dead bark tissue, before its peeling, falling off the tree and decomposing in the soil (which could be followed again by the uptake of ^{137}Cs from the soil solution via the root system).

The results shown in this study suggest that the ^{137}Cs activity distribution in plant tissues is different for each individual tissue and is directly related to the level of physiological activity in that particular tissue. This is manifested in the radial distribution by the increase in the value of activity with decreasing age (the highest activity was recorded in the youngest rings), while in the vertical distribution it is manifested only in the aboveground part through increasing activity with tree height.

The level of ^{137}Cs activity in the samples of chestnut rings in this study was highest in the highest part of the trees, as well as in the common birch in the studies conducted by [14] where the sampled birch trees were approximately the same age as the chestnut trees.

Investigating the radial and vertical distribution of ^{137}Cs in the trees of Japanese pine (*Pinus densiflora* Siebold and Zucc.) and Japanese oak (*Quercus serrata* Murray) after the nuclear disaster in Fukushima [18], it was found that ^{137}Cs activity for both species was highest in the outer bark, followed by the inner bark and rings (hardwood and softwood-heartwood). Mean values of ^{137}Cs concentrations measured on Japanese oak trees (*Quercus serrata* Murray) were 9,400 Bq kg^{-1} for the outer bark, 340 Bq kg^{-1} for the inner bark, 72 Bq kg^{-1} inside the softwood, and 12 Bq kg^{-1} in hardwood. The results of measuring ^{137}Cs activity levels very soon after the Fukushima accident, in some tissues exceed the measured ^{137}Cs activities in this study by several orders of magnitude. It must be emphasized that the research of Ohashi et al. [18], in which ^{137}Cs activity level in the forest ecosystem was measured, was conducted immediately after the Fukushima accident, in contrast to this study which was conducted over the past fourteen years, and the conditions affecting the results were not identical. Nevertheless, the results of analyses by Ohashi et al. [18] on Japanese oak samples are comparable to the results obtained in this study on chestnut samples because they indicate a similar vertical and radial distribution of ^{137}Cs in the tissues of two different but related tree species, (angiosperms, belonging to the same *Fagaceae* family).

In the outer bark of Japanese oak, a significantly higher concentration of ^{137}Cs was measured in the upper part of the trees than at the base of trees, while in this study the vertical distribution was distributed differently, i.e. a higher level of ^{137}Cs activity in samples of dead (outer) chestnut bark was measured at the base of trees and the smallest at the highest part of the tree.

In contrast to the ratio of the values of the vertical distribution of ^{137}Cs, the radial level of ^{137}Cs activity in the tissues of chestnut trees in this study (as follows: dead (outer) bark > inner (live bark) > rings) was the same as in the Japanese oak survey [18].

Studies performed on samples of chestnut trees in this study showed higher values of ^{137}Cs concentration in bark samples compared to those in rings, and similar results were found by Zhiyanski et al. [19], based on their research. They found higher ^{137}Cs activity in the bark of pedunculate oak trees (deciduous tree species from the same family as chestnut), compared to newly formed tree biomass (rings), and concluded that in the years after radioactive entry into the environment, ^{137}Cs activities may change bark structure due to physiological and ecological influences. The accumulation of ^{137}Cs in tree bark is mainly the result of direct adsorption after the Chernobyl disaster and persists on the outer bark for many years.

After the accident in Fukushima, Japan, research was also conducted on the tissues of the Japanese chestnut (*Castanea crenata* Siebold and Zucc.) (leaves, fruits, bark, wood) and larvae of *Curculio sikkimensis* Heller (*Coleoptera: Curculionidae*), which develop in Japanese chestnut [20]. Samples of this study were collected on three occasions (hedgehogs, fruits and larvae in October 2013, leaves and soil samples in November 2013, and bark and rings of trees in February 2014). The results of the analysis of leaf, bark and ring samples of that study showed a similar range of ^{137}Cs distribution in Japanese chestnut tissues as recorded in this study on chestnut samples in the winter. In the fruit nucleus of the Japanese chestnut higher ^{137}Cs activities were recorded in the central part than in the outer part and hedgehogs. As a possible reason, Sasaki et al. [20] hypothesized that a relatively large amount of nutrients is stored in the central part of the nucleus (cotyledon) to feed the fruit and in this case the beetle larvae also. It is appropriate to point out that in this study a higher level of ^{137}Cs activity was measured in ripe chestnut fruits sampled in winter compared to samples during vegetation, with lower activity recorded in fruit (and higher in hedgehogs) in winter, while in the vegetation period it was the other way around. It can be concluded that the results of this study in fruit and hedgehog samples do not coincide with those found by Sasaki et al. [20] for the winter period, but on the other that Sasaki et al. [20]

covered only part of the variability in the distribution of ^{137}Cs activity in Japanese chestnut tree tissues, given the significant influence of the time of year on that distribution observed in this study. It can also be established that they conducted sampling for analysis on several occasions in the same vegetation period, in contrast to the sampling conducted as part of this study when for each vegetation period it was sampled on one occasion. Accordingly, it is assumed that both the time and method of sample collection can affect the results obtained. Given the sampling time, very soon after the accident in Fukushima, Sasaki et al. [20] measured a 10-fold higher level of ^{137}Cs activity in the bark of the Japanese chestnut (*Castanea crenata* Siebold and Zucc.) than in tree rings, which coincides with this study in both vegetation periods. Such results indicate that the deposited ^{137}Cs from the bark penetrates the inner part of the tree (rings) faster than the tree absorbs it from the root system.

With this study, the measured distributions of ^{137}Cs in chestnut wood had the highest level of ^{137}Cs activity in tissue samples collected from the highest tree heights (annual apical shoots and leaves) which is comparable to the data of Fesenko et al. [21] who also recorded the highest levels of ^{137}Cs activity in annual branches of Scots pine and birch in Russia. They explained their results by the fact that the tree canopy played the role of a natural ^{137}Cs filter taken over from the root when radionuclides are transported from the root system to the xylem (tree conduction system) and then to the leaves or needles, and the main ^{137}Cs was stored in a newly formed annual ring. Such a transfer process leads to a decrease in ^{137}Cs activity concentration in the xylem, which is more pronounced in the lower part of the tree and decreases to the level of 2/3 of the tree height, because above that height the tree diameter decreases towards the top of the tree, i.e. the cross-sectional area through which radionuclides are transported is reduced. This leads to an increase in concentrations from the middle to the top of the tree and this effect is obviously more pronounced for higher trees.

In order to better understand and possibly clarify the long-lasting presence of ^{137}Cs in the different forest ecosystems, this investigation, in addition to measuring ^{137}Cs activity levels in chestnut tree tissues and soils with sampled trees, included ^{40}K activity levels to try to clarify the relations between ^{40}K and ^{137}Cs activities in trees and soil. The highest values of ^{40}K concentrations in chestnut samples were measured in the youngest tissues, and in older tissues (rings) they were constant values, compared to ^{137}Cs whose activities were measured above the detection limits only in rings of trees aged 1-5 years.

It is documented that ^{137}Cs transfer depends on soil characteristics, especially organic matter content, texture and cation exchange capacity. Since clay soils tend to accumulate ^{137}Cs and its sorption is conditioned by clay minerals, if the proportion of clay in the soil is higher, the activity of ^{137}Cs in the soil is increased, too [9, 22]. The K^+ content in the soil can cause the breakdown of the expanded interlayers [23], due to the ^{137}Cs binding within the interlayers and its blocking and inaccessibility to transmission processes.

Previous studies of the relationship between ^{40}K and ^{137}Cs in soil have been mainly focused on measurement and determination of the distribution and level of ^{137}Cs activity in soil after the Chernobyl and Fukushima tragedies [9, 24-29]. Their importance was strongly connected to the remediation of the consequences that occurred after the entry of ^{137}Cs into the environment and to taking appropriate measures for safer food production and animal husbandry, but also for forest ecosystems. Kaunisto et al. [28] conducted research in forest ecosystems of Scots pine (*Pinus sylvestris* L.) and in soils in these habitats, and found that in a forest ecosystem that lacks potassium in the soil, Scots pine trees absorb more ^{137}Cs from the soil than under normal conditions, i.e. after treatment with fertilizers, they measured a significantly reduced intake of ^{137}Cs into tree tissues.

3.4 Concentration of ^{40}K and ^{137}Cs, Clay Content, Amount of Organic Matter and Potassium Concentration in Soil

The results of measuring the concentration of ^{40}K and ^{137}Cs in the soil are shown in Tab. 3.

Table 3 Activities of ^{137}Cs and ^{40}K (Bq kg^{-1}) in soil (composite sample 0 - 5 cm and 0 - 15 cm) in the vicinity of chestnut felled near Petrinja (samples from December, 2 2003 and April, 14 2017)

Composite soil sample	^{40}K	^{137}Cs
0 - 15cm (2003)	516.3 ± 12.6	93.3 ± 1.4
0 - 15 cm (2017), (on 01/07/2003)	554.0 ± 63.0	185.0 ± 19.0
0 - 15 cm (2017), (on 01/07/2016)	554.0 ± 63.0	137.0 ± 14.1
0 - 5 cm (2017), (on 01/07/2003)	560.0 ± 63.5	192.0 ± 19.7
0 - 5 cm (2017), (on 01/07/2016)	560.0 ± 63.5	142.0 ± 14.6

Additional analysis of composite forest soil samples from depths of 0 - 5 cm and 0 - 15 cm from the Petrinja area are shown in Tab. 4.

Table 4 Results of soil analysis for clay content, plant-accessible potassium (mg K_2O / 100 g soil) and organic matter content (sample from April, 14 2017)

Depth of sampling	% of clay (< 0.002 mm)	AL-K_2O (mg/100 g soil)	% of organic matter
0 - 15 cm	21.22	12.42	9.39
0 - 5 cm	12.30	21.73	8.10

In soil samples from the Petrinja area (0 - 5 cm), a higher level of ^{137}Cs activity was measured where ^{137}Cs was biogeochemically retained almost entirely in the first 5 cm in the soil than at a depth of 0 - 15 cm from the same location (Tab. 3). It leads to the conclusion that soil characteristics have a crucial role in ^{137}Cs transfer in the forest ecosystem, especially given the share of clay in soils (which increases the activity of ^{137}Cs in soil due to adsorption of ^{137}Cs on clay minerals) and the amount of organic matter (Tab. 4).

4 CONCLUSIONS

The results of research on the distribution of ^{137}Cs and ^{40}K in chestnut trees (*Casanea sativa* Mill.) from the Banovina region on three occasions between 2003 and 2016, which included samples of tree rings from three heights (separated into bark and rings), roots, leaves, peak shoots, fruits and soil samples with fallen trees, and which were obtained after laboratory processing, statistical processing and interpretation indicate the following conclusions.

Throughout the investigated period (from 2003 to 2016) the distribution variability of ^{137}Cs in chestnut trees

decreased but will affect the environment for many years to come. The use of wood as a raw material and its by-products is inevitable, and the measured results indicate that there is no danger of an increased radiation dose. This was due to radioactive decay (about 26% in all tissues from 2003 to 2016). Also, it was due to the gradual elimination of ^{137}Cs from tree tissue, especially through dead bark and leaves, which caused a decrease in ^{137}Cs activity up to 92% in the dead chestnut bark (relative to the normalized values, after zeroing of the impact of radioactive decay). In winter, the highest activities were recorded in the top shoots, leaves, fruits and hedgehogs, while in the vegetation season, the highest activities were in the top shoots and dead bark. From this it can be concluded that the dominant ^{137}Cs elimination pathways in chestnut are tissues that the tree sheds in a relatively short interval in autumn. A decrease in ^{137}Cs concentrations was observed in the rings with increasing age of the rings. The highest level of ^{137}Cs activity was measured in the highest part of the trees. In chestnut habitat soils, ^{137}Cs was biogeochemically retained almost entirely in the surface layer (0 - 15 cm). Significant increased values of activity were measured in samples of the youngest and physiologically most active parts of trees, compared to the least physiologically active where concentrations were lower, which indicates the possibility of considering the spatial distribution of ^{137}Cs in tree tissues not only from the perspective of radioactive contamination of anthropogenic origin but also by understanding ^{137}Cs as a tracer of the level of physiological activity. Concentrations of ^{137}Cs and ^{40}K from the same samples in chestnut were not statistically significantly correlated (neither for all tissues, nor for individual tissues separately). With respect to the fact that no statistically significant negative correlation was observed (and a significant positive correlation can still be interpreted by a random process), these findings suggest that the chestnut trees (*Casanea sativa* Mill.) do not differ caesium and potassium, as two homologous elements.

This research was the first of its kind in Croatia primarily focused on the tissues of edificatory tree species. Obtained results contribute to better understanding of the fate of ^{137}Cs that entered the tissues of mentioned tree species in the forest ecosystem, as well as its time and spatial distribution which completes the picture of the biogeochemical cycle of ^{137}Cs in the environment (especially in forest trees).

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