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Cesium-137 contamination of oak (*Quercus petrae* Liebl.) from sub-mediterranean zone in South Bulgaria

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ABSTRACT

This study focuses on the cesium-137 (^{137}Cs) contamination in grass and in different compartments of oak trees growing in ecosystems, located in the zone with sub-mediterranean climate in South Bulgaria, characterized with high summer temperatures, low precipitation and often periods of drought. In 2008, three experimental sites – PP1, PP2, PP3 – were sampled in oak ecosystems from Maleshevska Mountain at 900 m above sea level. Samples from grass species and oak tree leaves, branches with different diameter, wood disks and bark were analyzed for ^{137}Cs activity with γ-spectrometry. The soil-to-plant transfer factor (TF) values for ^{137}Cs were estimated differentiating different tree compartments. Our findings showed relatively high activity concentrations of ^{137}Cs in oak trees even 22 years after the Chernobyl accident. The grass under oak was less contaminated compared with the oak trees. The different organs of oak trees could be distinguished according to the ^{137}Cs contamination as follows: bark > branches ($d < 1$ cm) > leaves > branches ($d > 3$ cm) > wood. The relatively higher contamination of bark compared with the new-formed biomass suggested that a significant part of ^{137}Cs was accumulated as a result of direct adsorption at the time of the main contamination event. The TF values obtained and the presence of ^{137}Cs in the branches, leaves and in the wood formed after 1986 confirmed that 22 years after the contamination, the main mechanism of ^{137}Cs entrance in tree biomass was the root uptake.

1. Introduction

Due to the accident with the NPP "Chernobyl" in 1986, the environment in Europe was strongly contaminated with artificial radionuclides. Recently scientific efforts were directed toward better comprehension of the transfer phenomena of radionuclides in different types of terrestrial ecosystems such as agro-ecosystems, pastures and forests (Desmet and Myttenaere, 1988; Bunzl and Kracke, 1988; Bunzl et al., 1989; Ronneau et al., 1991; Fesenko et al., 2001; Soukhova et al., 2003; Steiner, 2004; Goor and Thiry, 2004). Good knowledge about the behaviour of radioactive elements in forest ecosystems is of great importance: the understanding how these elements are accumulated, distributed and changed in different components of ecosystem is obligatory to risk assessment of both humans and the environment.

The ^{137}Cs is a dangerous radionuclide for humans in terms of its relatively long half-life and its high mobility in biological systems. This fact is attributable to the persistence of ^{137}Cs in all compartments of forests, pastures and natural meadows. In the ^{137}Cs cycle, both the soil and vegetation are major reservoirs of this radionuclide (McGee et al., 2000). The ^{137}Cs transfer through the food chain in agricultural ecosystems is relatively well studied (Cougthry and Thorne, 1983; Nisbet and Woodman, 2000; Shaw et al., 2007). Meanwhile the study of ^{137}Cs transfer in soil–plant system of forests, meadows, tundra, etc., is still in progress. The high scientific interest to study the ^{137}Cs contamination in forest ecosystems is provoked by the fact that forests are complex environments with great capacity to intercept and to retain radionuclide deposition for a long time (Adriano et al., 1981). There is also a concern to restrict radionuclide transfer out of the polluted forests (Prister et al., 1991). Tikhomirov

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and Shcheglov (1994) established that the ^{137}Cs contamination in forests is up to 30% higher compared with agro-ecosystems.

Knowledge about the long-term behaviour of ^{137}Cs in forests is needed in order to predict the future contamination levels of wood products from the affected areas (Fogh and Andersson, 2001). Cesium-137 has a high level of mobility within the plants, and in general, the activity concentrations of ^{137}Cs was much higher in the fresh parts of the tree (needles, leaves and twigs) than in the core wood of pine, oak and birch studied in the strong polluted region of Briansk (Fogh and Andersson, 2001). Soukhova et al. (2003) established different patterns of ^{137}Cs distribution in coniferous (*Pinus sylvestris*) and deciduous (*Betula pendula*) trees and suggested that possible explanation for this is a different radial ray structure between the tree species. For a given species the ^{137}Cs radial distribution can be influenced by the age of tree and site characteristics, therefore, more information about this issue is needed for further studies.

The ^{137}Cs transfer in ecosystems from Bulgaria before the Chernobyl accident in 1986 was studied by Raikov (1978) and Semerdjieva and Dimchev (1983). Several investigations on the soil-to-plant transfer and distribution of ^{137}Cs due to its fallout after the Chernobyl accident in 1986 were performed in the forest ecosystems in mountainous regions of Bulgaria (Klein et al., 1994, 1995; Yovtchev et al., 1997; Lucot et al., 1998; Sokolovska et al., 2006; Zhiyanski et al., 2005, 2006a, 2006b, 2008, 2009 and references therein). The oak ecosystems (*Quercus petrae* Liebl.) located in the south part of Bulgaria are less investigated with respect to radio-ecological studies (Sokolovska et al., 2006). These ecosystems are particularly interesting because the zone characterizes with Mediterranean climatic influence and relatively high level of ^{137}Cs contamination in soils.

This study focuses on the ^{137}Cs activity concentrations in grass and in different organs of oak trees from forest ecosystems in South Bulgaria. The observation of the cross-section of the wood allows distinguishing the different zones corresponding to the growing periods before and after the Chernobyl accident. The analysis of the different parts of the trees allowed to evaluate the spreading of ^{137}Cs in the tree and to determine different soil-to-plant TFs.

2. Materials and methods

2.1. Study region

The study ecosystems were formed by oak (*Q. petrae* Liebl. with single participation of *Fagus sylvatica* L.) and located in Maleshevska Mountain at 900 m a.s.l. The region is a part of the Bulgarian sub-mediterranean region, which actually represents the most northern part of the zone with real Mediterranean climate. The summer season often has persistent drought and this region has the lowest amount of precipitation for the country (Dimitrov, 1986).

The studied oak forests had the following characteristics: tree species – *Q. petrae* Liebl.; mean age = 100 years; origin – natural, high-stand; dominant species in undercover include *Sorbus torminalis* (L.) Crantz., *Mespilus germanica* L., *Festuca arundinacea*, *Trachystemon orientale*, *Euphorbia amygdaloides*, *Lactuca muralis*, etc. In oak ecosystems three experimental sites – PP1, PP2, PP3, with an area of 0.5 ha each, were chosen and their components were sampled in order to investigate the heterogeneity of spatial ^{137}Cs distribution and contamination of vegetation.

2.2. Soil and forest floor

The soils are *Dystric Cambisols* with soil profile from type $AhB_1B_2B_3/C$, underlain by granites and granite–gneisses. The brown forest soils have acid pH (pH in H_2O varied from 4.7 to 5.7), sandy–loam texture and low organic carbon content (from 2.1% in Ah soil horizon decreases to 0.6% in deeper soil horizons) (Zhiyanski et al., 2009). One representative soil profile (1:1.5:1 m) was prepared in each site next to the model oak tree according to the recommendations of Takenaka et al. (1998). The forest floor layers Aol and Aof + h were sampled in 5 repetitions per site with a 25/25 cm frame, and one composite sample of each layer was formed. Samples from soil depth layers 0–5 cm, 5–10 cm, 10–15 cm, 15–20 cm, 20–25 cm and 25–30 cm, respectively, were collected. In the laboratory the soil samples were ground to powder and analyzed for chemical characteristics and ^{137}Cs activity.

The ^{137}Cs distribution and its migration in the soil system of the experimental sites were reported in previous papers and could be summarized duly that the mean total ^{137}Cs activity in the upper 0–30 cm soil layer was 186 ± 4 Bq kg^{-1} (for the surface 0–5 cm the ^{137}Cs activity was 103 ± 7 Bq kg^{-1}) and the forest floor was also contaminated, the mean total ^{137}Cs activity was 146 ± 2 Bq kg^{-1} (Sokolovska et al., 2006; Zhiyanski et al., 2009). These results were used for estimation of the radionuclide TF in the present study.

2.3. Biomass

The biomass of aboveground oak trees was studied in detail using the methods applied in forestry. In each experimental site one representative model tree was identified (method of Nedyalkov et al., 1983; Schuetz, 1999), and named model tree 1, 2 and 3, respectively. The wood disks of each model tree were cut as follows: in the base of the trunk and at distances of 1.3, 2, 4, 6, 8 and 10 m upward from the base. The bark at each distance was carefully separated, ground and analyzed. The wood disks were dated and the year of Chernobyl accident, 1986, was fixed. Then the wood was separated at two parts – internal disk – wood formed before 1986 and external disk – the wood formed after 1986. Two samples per disk (one for internal and one for external) were obtained by drilling, ensuring a representative age cross-section for each part, collecting approximately 50–70 g. The branches were differentiated by class of diameter (d) as follows: with $d < 1$ cm, 1–3 cm, $d > 3$ cm. Then the bulk sample per each class of diameter for model tree was prepared, ground to powder and analyzed for ^{137}Cs activity. The leaves from each model oak tree were collected in a single sample. The grass was sampled in total of 15 plots with a 1×1-m frame. For analyses the composite samples from 5 repetitions were used. In total, there were 3 samples of grass, 3 of oak trees leaves, 9 of branches, 21 of wood disks (with a total of 42 wood samples: 21 for internal and 21 for external parts) and 21 samples of bark, analyzed in 5 repetitions.

All samples were transported to the Laboratory of Soil Science in the FRI-BAS, Sofia, then dried, ground and prepared for further analyses. The ^{137}Cs activity in samples was measured with γ-spectrometry in the Institute of Nuclear Research and Nuclear Energy – BAS and in the National Centre for Radiobiology and Radiation Protection-MH. The activities of ^{137}Cs in the collected samples were measured in accordance with International Standard IEC 61452 (1995–09) Nuclear instrumentation – Measurement of gamma-ray emission rates of radionuclides – Calibration and use of germanium spectrometers. The determination of gamma emitting radionuclide by gamma-spectrometry characterized by the Lower Limit of Detection for tested soil and vegetation materials for ^{137}Cs < 1 Bq kg^{-1}.

Transfer factor has been used for many years to predict concentrations of radionuclides that could be expected in food crops after accidental releases of radionuclides into the environment. The concentration of the radionuclide (i) in the plant or in the part of the plant (C_i^p) [Bq kg^{-1} dry mass] is linearly related with the concentration of this radionuclide in the rooting zone or in the surface soil layer (C_i^n) [Bq kg^{-1} dry soil] (Ehlken and Kirchner, 2002).

$$C_i^p = TF_i * C_i^n \quad (1)$$

Proportionally the concentration TF_i in the equation (1) is the transfer factor from soil to plant.

2.4. Statistical analysis

The data obtained about the ^{137}Cs activity concentration were grouped and summarized according to the experimental sites and different studied compartments. The paired sample t-test was applied to compare the mean scores of two groups on a given variable. The acceptance of hypotheses was on the basis of the level of significance less than 0.05. Pearson correlation analysis was applied for establishment of correlations between variables. Because TF data are usually skewed, log transformation was applied in statistical analysis. The SPSS 13.0.1 Version for Windows was used.

3. Results

3.1. Cesium-137 activity in grass vegetation and in different organs of oak trees

The ^{137}Cs activity concentration determined in grass under the oak stands was very low and varied from 0.1 to 1.3 Bq kg^{-1} (Table 1). The ^{137}Cs contamination in the oak leaves ranged from 3.2 to 4.3 Bq kg^{-1} (Table 1), with a mean of 3.6 ± 0.80 Bq kg^{-1}. For the branches with $d = 1$–3 cm, the ^{137}Cs activity concentration was below the LLD and are not included in the table (Table 1). The ^{137}Cs in the newly formed branches ($d < 1$ cm) ranged from 1.4 to 4.6 Bq kg^{-1}. For the branches with $d > 3$ cm, ^{137}Cs was ranged from 1.2 to 6.8 Bq kg^{-1}. As a whole the contamination of young branches (2.6 Bq kg^{-1}) was lower than the mean detected activity of older

Table 1
Cs-137 activity (Bq kg^{-1}) in different compartments of oak trees in studied area.

Site/tree No.	Grass under oak	Oak leaves	Oak branches		Oak bark	Oak wood	
			<1 cm	>3 cm		<1986	>1986
PP1/tree 1	1.2 ± 0.9	4.3 ± 0.8	4.6 ± 1.0	4.0 ± 0.6	6.5 ± 0.2	0.8 ± 0.2	1.0 ± 0.2
PP2/tree 2	1.3 ± 1.0	3.4 ± 1.0	1.9 ± 0.6	1.2 ± 0.7	12.1 ± 0.2	0.8 ± 0.2	0.8 ± 0.2
PP3/tree 3	1.3 ± 0.1	3.2 ± 0.6	1.4 ± 0.6	6.8 ± 0.9	12.0 ± 0.1	1.0 ± 0.2	1.5 ± 0.9
Mean	1.3 ± 0.7	3.6 ± 0.8	2.6 ± 0.7	4.0 ± 0.8	10.2 ± 0.2	0.9 ± 0.2	1.1 ± 0.4

ones (4.0 ± 0.8 Bq kg^{-1}), while the leaves (3.6 Bq kg^{-1}) had similar contamination to the older branches.

The activity concentration of ^{137}Cs in oak wood formed after 1986 (external disks) was higher than in the internal cylinder (before 1986). Comparatively, the ^{137}Cs contamination of wood was lower than in the other parts of oak trees and varied from 0.8 to 1.5 Bq kg^{-1}. The ^{137}Cs in the external wood rings were studied with respect to the distance from the base of the trunk (Fig. 1.). The ^{137}Cs activity concentration varied from 0.9 to 2.1 Bq kg^{-1} and the highest values were detected at the base of the trunk.

The ^{137}Cs was at highest concentrations in the bark of studied oak trees. The ^{137}Cs activity concentrations in bark varied from 2 Bq kg^{-1} to 23 Bq kg^{-1} and above 6 m from the base of the trunk there was a decrease in ^{137}Cs activity (Fig. 2). The bark at breast height (1.3 m) was most contaminated – 18–23 Bq kg^{-1}.

The cesium-137 contamination of grass was significantly lower than in oak trees compartments (Table 2). The branches with different diameter and the internal and external wood disks are not significantly different with respect to activity concentrations of ^{137}Cs.

3.2. Soil-to-plant transfer factor for ^{137}Cs

The estimated TF for the grass in the oak forests was low (mean = 0.007 ± 0.004) (Table 3). The TF corresponding to leaves was about 0.019 ± 0.005 (Table 3). The estimated TFs for branches were significantly higher compared with TF values for other compartments of studied oak trees. The ^{137}Cs TF values for the external and internal wood disks are quite similar (0.004–0.008) without significant difference and the TFs determined for the whole wood part were low (0.005 ± 0.004). These TF values were the lowest determined.

4. Discussion

According to the data reported by Petkov et al. (1976), the average ^{137}Cs activity concentration for grass species in Bulgaria before the Chernobyl accident was estimated at 2.1 ± 0.1 Bq kg^{-1}, which shows that the ^{137}Cs contamination of grass species under oak in 2008 is very low. Kühn et al. (1984) informed that the soil-to-grass TF values for ^{137}Cs in Germany before 1986 were from 0.01 to 0.1, which are quite similar to data obtained in the present study. Papastefanou et al. (1999) reported for high variation in TF for ^{137}Cs toward grass species in Northern Greece from 0.002 to 7.42 (mean 0.20) and found no correlation between TF values and the time since contamination. The TFs here are within the range reported in other studies in Bulgaria where TFs for ^{137}Cs from soil to grass vegetation were 0.002–0.02 (Raikov, 1978; Djingova and Kuleff, 2002).

It has been shown (Antonopoulos-Domis et al., 1997) that ^{137}Cs concentration C_m (Bq kg^{-1}) of leaves at year $m \geq 1$ after the Chernobyl accident is firstly due to translocation of ^{137}Cs from the tree reservoir and the second term is due to root uptake (Antonopoulos-Domis et al., 1990). It has also been proven experimentally (Antonopoulos-Domis et al., 1996) for fruits and leaves of fruit trees. In the first four years following the Chernobyl accident only translocation is important for the contamination of fruits and for the years after 1991, root uptake is the dominant mechanism. For leaves from oak trees, Clouvas et al. (2007) found that at least after 1992, the dominant mechanism of ^{137}Cs contamination of leaves was due to root uptake. In this case, the TF from soil to leaf has meaning. On the contrary the TF concept is meaningless in cases where the root uptake mechanism is not important. For that reasons we do not include TF values for the bark in this study and the TF values obtained are valid only for tree compartments shown in Table 3.

Despite the differentiation of oak branches by diameter, the contamination of branches was relatively higher (up to 6.6 Bq kg^{-}1) than the contamination in leaves and wood. Therefore, a higher TF for branches was determined – between 0.02 and 0.05 (mean 0.04) (Table 3).

The comparative analysis showed that at 1.3 m the external wood was less contaminated (0.6–1.6 Bq kg^{-1}) than the bark

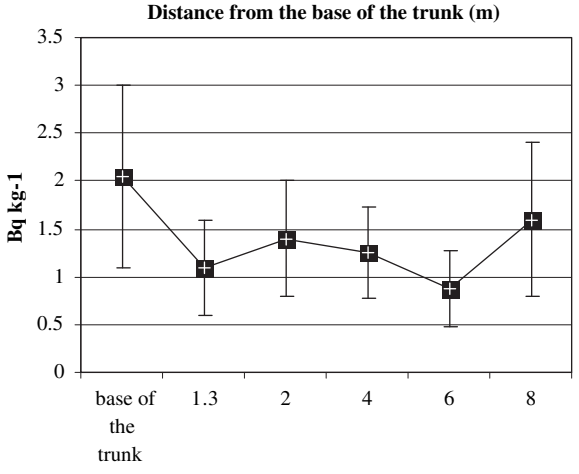

Fig. 1. Cesium-137 activity concentrations in the external wood (formed after 1986) of oak trees at different sections.

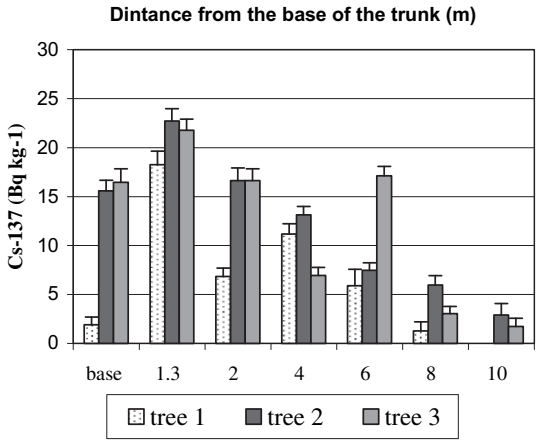

Fig. 2. Cesium-137 activity concentrations (Bq kg^{-1}) in the bark of oak trees at different distances from the base of the trunk.

Table 2
Paired sample *t*-tests applied to data obtained for various compartments of oak trees.

	Leaves	Branches < 1 cm	Branches > 3 cm	Bark	wood < 1986	wood > 1986
Grass	[b]	[a]	[a]	[b]	[a]	n.s.
Leaves		[a]	n.s.	[b]	[b]	[b]
Branches < 1 cm			n.s.	[b]	[a]	[a]
Branches > 3 cm				[b]	[b]	[b]
Bark					[b]	[b]
Wood < 1986						n.s.

n.s.: not significant.
[a] Mean scores are significantly different at the 0.05 level.
[b] Mean scores are significant different at the 0.01 level.

(18–23 Bq kg^{-1}). The bark at breast height (1.3 m) was most contaminated – 18–23 Bq kg^{-1}. Barci-Funel et al. (1995) analyzed three different coniferous species: Scot's pine, Norway spruce and Larch. The activity concentration of ^{137}Cs in different tree organs – leaves, branches and bark was 3–17 times higher than the contamination of internal wood disk, which showed an "external accumulation". The same authors established that the ^{137}Cs in the bark was 1–2 times higher than in the wood as well as 3–17 times higher than the needles. In other study on Scot's pine and Norway spruce, McGee et al. (2000) published similar results, where the activity of ^{137}Cs in the bark was only just five times higher. Fogh and Andersson (2001) have obtained similar results for Scot's pine, birch and oak. They established that the ^{137}Cs activity in the active growing organs (leaves, needles and bark) was times as much higher than this in older organs (such as wood).

Our data show that the ^{137}Cs activity in the bark is 15 times higher than that in the wood and up to 9 times higher compared with other tree organs. Moreover the ^{137}Cs was relatively uniformly distributed in the different sections up to 6 m distance from the base of trunk. The ^{137}Cs activity concentrations were high in the base of the trunks, and at 2 and at 4 m distance. After 6 m a decrease in ^{137}Cs contamination in the bark was detected. Bark has a relatively large concentration of ^{137}Cs, obviously due to absorption from direct wet deposition. Most, if not all, of ^{137}Cs in the bark, is not available for translocation to the biomass of other parts in the tree. As root uptake is not the dominant mechanism for ^{137}Cs contamination in the bark, the TF concept for bark is meaningless. It could be supposed that there was a probability of ^{137}Cs to be additionally deposited on the bark through foliar leaching and/or stemflow after main contamination, but in a very low concentration, which could be neglected. Additional studies on this issue are needed in order to support this assumption.

The measurements of ^{137}Cs contamination of wood in the internal and external disks showed that the mean ^{137}Cs contamination in the external disks was quite similar to the radionuclide concentration of internal disks. The wood rings formed before 1986 are contaminated mainly through the diffusion from the outer rings (Antonopoulos-Domis et al., 1990). Because of the fact that the diffusion is a very slow process, most of the rings in the inner part of the trunk are still not contaminated and in some of our samples ^{137}Cs was under the LLD. Additionally it could be accepted that the wood rings formed before 1986 were polluted through the processes of root uptake from previous depositions (nuclear tests).

Table 3
Transfer factors for ^{137}Cs in grass and different oak tree compartments.

Site/tree No.	Grass	Leaves	Branches	Wood
PP1/tree 1	0.006 ± 0.005	0.023 ± 0.004	0.046 ± 0.003	0.009 ± 0.001
PP2/tree 2	0.007 ± 0.005	0.018 ± 0.005	0.016 ± 0.005	0.007 ± 0.001
PP3/tree 3	0.007 ± 0.001	0.018 ± 0.004	0.046 ± 0.005	0.006 ± 0.005
Mean*	0.007 ± 0.004	0.019 ± 0.005	0.036 ± 0.015	0.005 ± 0.004

* The paired sample *t*-test applied showed significant difference between mean scores of all tested variables.

The external disks (after 1986) are polluted by the ways of root uptake from the superficial soil horizons (Ah and B_1). The similarity in data obtained for the internal and external disks (not significant difference was determined) as well as the low contamination in grass supposed that the bigger part of ^{137}Cs in the superficial soil is strongly fixed and the content of bioavailability forms is lower. This is attributed by the low organic matter content in the soils.

Nisbet and Woodman (2000) presented the typical variation of TF for cereals (0.0004–0.25), vegetables (0.08–1.7), and grass species (0.01–1.0). The mean value for soil-to-grass vegetation TF for ^{137}Cs in our study is 0.007, which is significantly lower compared with this value for different compartments of oak trees. Gommers et al. (2005) also informed that the TF from soil to oak trees wood varied from 0.001 to 0.457, and our results are within these limits. Our TF values for ^{137}Cs soil to different oak tree compartments ranged from 0.01 to 0.38 and the results are comparable with these reported by Al-Oudat et al. (2006) for oak trees in arid regions, which suggests a relatively higher level of ^{137}Cs transfer toward oaks under these climatic conditions. The data obtained showed that the ^{137}Cs transfer was clearly expressed in the basis of the trunk. The values of TF and the presence of ^{137}Cs in the branches, leaves and in the wood formed after 1986 (even the lower ^{137}Cs active concentrations) confirmed that 22 years after the contamination, the main mechanism of entrance and contamination of new-formed tree biomass is the root uptake.

The ^{137}Cs contamination of oak tree organs decreases in the order:

$$\text{Bark} > \text{young branches}(d < 1\ \text{cm}) > \text{leaves} > \text{old branches}(d > 3\ \text{cm}) > \text{wood}$$

5. Conclusions

The ^{137}Cs contamination of oak trees from the zone with Mediterranean influence of South Bulgaria, 22 years after the Chernobyl accident, was relatively high. The grass under oak stands was less contaminated compared with the aboveground trees. The different organs of oak trees can be distinguished according to the level of contamination with ^{137}Cs as follows: bark > young branches ($d < 1$ cm) > leaves > older branches ($d > 3$ cm) > wood. The higher contamination of bark compared with the newly formed biomass supposed that a significant part of ^{137}Cs was accumulated as a result of direct adsorption at the time of main contamination. The presence of ^{137}Cs in the branches, leaves and in the wood formed after 1986 confirms that 22 years after the contamination, the main mechanism of entrance and contamination of tree biomass is the root uptake. The corresponding TFs have been computed.

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