Forest litter as an indicator of radioactive pollution

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The experimental data on the content of radionuclide ^{137}Cs in the forest litter of Latvia forests after the Chernobyl accident are presented.

The investigation evaluates the role of forest stand structure in dissipation on the territory where the radionuclide ^{137}Cs has accumulated in the soil after the Chernobyl accident. The comparison of the concentration of ^{137}Cs in the upper layer of the soil was carried out in the pine, spruce and birch forests, and outside forest territories, as well – in Rucava (the influence zone of the Chernobyl accident) and in Taurene (the nonpolluted zone).

Inside the pine forest in Rucava the concentration of ^{137}Cs fluctuates within the limits of 108-724 Bq/kg, in the spruce forest 205-2270 Bq/kg, outside the forest territory the concentration was 15-30 Bq/kg.

In the similar forest in Taurene the dispersion of these indices is in the pine forest 42-157 Bq/kg, in the spruce forest 19-133 Bq/kg, outside the forest territory 3-19 Bq/kg.

These data confirm the great role of the coniferous forests in the adsorption of ^{137}Cs from the air, and its redistribution within the forest.

Key words: cesium-137, pollution, forest litter, pine, spruce, Chernobyl accident

Introduction

The accident of the Chernobyl Nuclear Power Plant changed the radiological situation acutely in many countries. Some countries were affected insignificantly, however, in others it became a national disaster. Nevertheless, in these countries that were submitted to the relatively slight radioactive pollution, it is necessary to conduct research on the pollution and its influence on the environment.

After the decay of the short- and medium-lived radionuclides, the radioactivity background is created mainly by radionuclide ^{137}Cs. Besides, the data on the pollution, knowledge about its vertical distribution in the soil, its mobility, its ability to accumulate in plants and living organisms are necessary (Frissel, 1993). For our study of the radioactive pollution we chose litter. This choice was made because the greatest accumulation of the products of the nuclear fission from the atmosphere is observed in the forest (Aleksahin, Narishkin,1972). The authors Narishkin, Romanov et. al. (1987) explored some Latvian forest reserves and stated that prior to the Chernobyl accident the concentration of ^{90}Sr and ^{137}Cs was among 37 and 370 Bq/kg in the needles of pines and in the soil. The partial results (the autumn of 1991) of the large survey of the radioactive contamination in the Polish forests, predominantly coniferous, are presented by Mietelski, Macharski et. al. (1994). The main part of Chernobyl cesium is still present in the upper layer (A_0) of the litter. Such a conclusion is strongly supported by the fact that for all samples, the ratio of ^{134}Cs to ^{137}Cs was observed to be higher in the A_0 layer than in A_1. The experimental data on the content of radioactive isotopes of Cs in individual components of Lithuanian pine biogeocenosis after the Chernobyl accident are presented (Styro, Tamulėnaitė, 1994; Butkus, Morkūnas et al., 1994). The period of the effective half-life of the radioactive isotopes of cesium in the pine biogeocenosis was equal to one year approximately. The radionuclides of cesium in the upper layer of the forest soil are found principally in the uninterchangeable forms increasing with the depth in the mineral part of the soil. Styro, Filistovičius (1994) deal with the determination of the migration parameters and the factor of the exponential penetration into the podzol sandy soil of the Curonian Spit (Lithuania) mixed forest. The migration and further penetration of ^{137}Cs into the soil in future will occur supposedly from the organic layer of the litter desorbing

the radionuclides from the high molecular compounds of the fulvic acids as the layer of the litter and wooden settling decompose. Mushrooms (Berzins, Leja et al., 1993; Mietelski, Jasinska et al., 1994), lichens and moss (Filipovics, Nikodemus, 1993), are good indicators of the radioactive pollution. The current levels in the lichens are 1-2 orders of magnitude higher than in the vascular plants (Bretten, Gaare et al., 1992) Investigating the influence of the stable cesium on the transfer factor of the radiocesium in the system litter-mushrooms the authors (Zagrodzki, Mietelski et.al., 1994) conclude that the stable cesium can be regarded either as the carrier of the radiocesium or their rival. The same would be true for the potassium. It is observed (Varskog, Neumann, Steinnes, 1994) that plants growing in the poor soil have a higher change of ^{137}Cs than these growing in fertile soil. Less than 10% of the total radiocesium in the soil is in the form available to the plants. Stable Cs and radioactive cesium in the solution are not in equilibrium. This probably is because of the fact that the stable Cs is bound in the lattice of the soil mineral particles to a large extent. It appears that about 5-20% of the ^{137}Cs activity originates from the nuclear testing period (data from Norway, 1987-1989). The investigators have paid a great attention to the migration of the radionuclides. Petrajev, Ovsjannikova et. al. (1993) conclude that the migration depends on soil propreties, the variety of the forms of the radionuclides and their transformation in the soil. The forms are classified in the following fractions: the water soluble, the exchangeable (water soluble+extract from 1M ammonium acetate solution), the mobile (exchangeable +extract from 1M hydrochloric acid), the fixed. The last one approximately comprises 98%. In the period of time the vertical migration is anticipated as a result of disaggregation. Clay minerals bind ^{137}Cs mainly in the soil. The average speed of the migration of ^{137}Cs is characterized by 0.19 cm/year, that is twice as low as ^{90}Sr (Romanov, Martyushov et. al., 1993). It has been stated that pollution by ^{137}Cs increases within the eroded landscapes in comparison to the adjacent accumulative plains regardless of the distance from the Chernobyl NPP (Kvasnikova, Stukin, 1993). The level of the radioactivity in the soil depends on the humus content in it (Chelmicki, Mietelski et. al., 1993).

The aim of the study was to obtain data on the migration, accumulation, and distribution of the ^{137}Cs contamination on the Latvian forest territories by using the nuclear spectroscopy methods. It was important to investigate experimentally:

1) the depth of the ^{137}Cs migration within 8 years after the Chernobyl accident,

2) the pollution level in the specific regions,

3) whether the pollution level differs depending on various tree species,

4) the relation between the number of samples analysed and the reliability level of the obtained results (at various probability levels).

Materials and methods

Special attention should be focused on the investigation of the pollution levels in the forest in relation to the various tree stands. The regions with different levels of the radioactive pollution were selected: Rucava in South-western Kurzeme that was passed over by the tail of the radioactive cloud of Chernobyl and Taurene in Vidzeme, outside the area influenced by Chernobyl.

The samples were collected in the summers of 1994 and 1995. For the investigation of the accumulation of ^{137}Cs in the forest litter the samples of the acknowledged standard size, that is, of 5 cm thick layers were gathered. The data on migration of ^{137}Cs was gleaned at various depths (5 levels: from 0 to 25 cm). The size of the sample areas was 50x50 m. The samples on both different areas of the investigation were gathered choosing the stands:

1) of similar age and height, etc.

2) of identical types of forests

3) growing in the similar type of soil,

4) of the level area.

The density of the upper layers of the soil (A_0, A_1 layers) has the significant negative correlation between the forest litter on the selected area; the ratio of the correlation -0.63 shows that the soil is looser at places where there is more forest litter. Therefore along with conventional unit of measure Bq/kg of ^{137}Cs we also used Bq/l.

The samples were dried and homogenized in the "runner mill". The measurements of the radioactivity of ^{137}Cs were made, using the semiconductor gamma-spectrometer with HPGe detector of 70 cm^3. The background of the radioactivity was minimized by using the 80 mm thick shield of the lead and copper plate. The samples were measured in the Marinelli beaker at the top of the detector, the volume of the sample was 1 liter. The certified standard of ^{137}Cs with the activity of 1040 Bq was used in the process. The measurement time was

Table 1. Valuation of tree stands.

Points of observation	Tree species	Stands age, vears	Site index	The mean height, m	The mean diameter, cm	Density	Yield, m^3/ha
The Rucava forest	Pine	55	II	17.0	21.7	0.76	182
	Spruce	50	Ia	19.0	20.0	0.77	255
	Birch	50	Ia	25.5	26.0	0.75	244
The Taurene forest	Pine	50	II	16.0	17.3	0.83	185
	Spruce	80	Ia	32.0	37.5	0.80	510
	Birch	60	Ia	28.0	28.5	0.72	286

selected for the statistical error of the results. It did not exceed 3-5%.

Results and discussion

While experimentally estimating the migration of ^{137}Cs in the forest soil layer up to 25 cm deep, it was established that even 8 years after the Chernobyl accident up to 95% of ^{137}Cs is concentrated in the upper 5 cm layer. It confirms other authors' (Romanov, Martyushov et. al., 1993; Butkus, Morkūnas et al., 1994) data that 90-95% of ^{137}Cs are located in the soil in fixed (insoluble) condition and its migration takes place very slowly.

The concentration of ^{137}Cs in the upper layer of the soil in the pine stands is compared to the amount of forest litter accumulated from May to September of 1995 at the location of samples (data collected by M.Laivins).

Figure 1 shows that the correlation between the relative quantity of ^{137}Cs in the forest litter and relative quantity of the needles is linear.

It enables us to conclude that thicker forests are more polluted with ^{137}Cs.

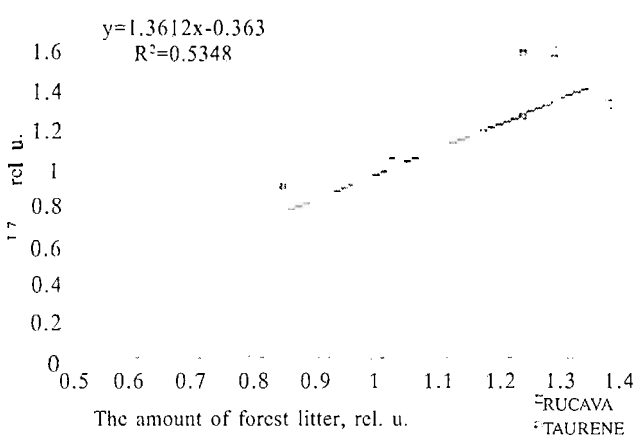

Fig. 1. The relationship between the concentration of ^{137}Cs in the upper layer of the soil and amount of the forest litter accumulated from May to September of 1995

In the forests of Taurene (in the nonpolluted zone unaffected by the Chernobyl accident) the concentration of ^{137}Cs in all forests, regardless of their structure, is significantly higher than in the arable land outside the forest (Fig.2).

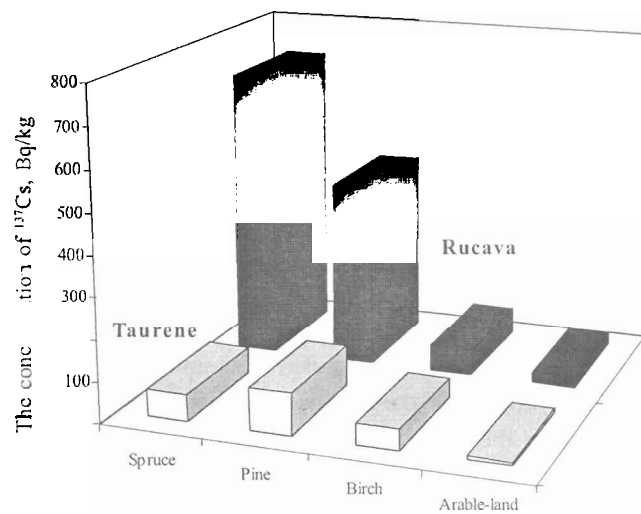

Fig. 2. Content of ^{137}Cs in forest litter, Bq/kg

The mean concentration of ^{137}Cs of the Taurene forest is 79 Bq/kg; but in soil volume – 57 Bq/l while outside the forest territories they are 9 Bq/kg and 13 Bq/l, respectively. The highest variations of ^{137}Cs are in the spruce forest — 19-133 Bq/kg; the variations in the concentration of ^{137}Cs both in pine and birch forests do not exceed these limits.

Significantly different indices are in the Rucava forests influenced by the Chernobyl accident (Fig.2.).

The highest ^{137}Cs content is established in the spruce forest; the mean values are 813 Bq/kg or 342 Bq/l. There are large differences in the concentration of ^{137}Cs in the same forest. The results obtained in two separate spots within 10 m distance differ from one another more than ten times: 205 and 2268 Bq/kg. Lower dispersion occurs in the pine forest: the concentration of ^{137}Cs in the samples analysed are within limits from

108 to 724 Bq/kg. The mean values are 446 Bq/kg or 196 Bq/l, that is significantly lower than in the spruce forest. The lowest concentration of ^{137}Cs is in the birch forest: on average 71 Bq/kg or 79 Bq/l. These indices do not differ significantly from the indices in the Taurene birch forest that was not influenced by the Chernobyl accident. It confirms the large abilities of adsorption of the crowns of the coniferous trees. It is should be remembered that during the Chernobyl accident the birch trees did not have leaves yet, therefore their ability of the adsorption was low.

On the territories outside the forest near Rucava the content of ^{137}Cs in the soil is on average 23 Bq/kg or 31 Bq/l, that is, approximately three times less than the radiocesium content in the birch forest, 20 times less than in the pine, and 40 times less than in the spruce forest. The results similar to these about the concentration of ^{137}Cs in the dunes and in the pine stand behind the dunes are presented by Butkus, Morkūnas et al. (1994). Until now, we did not have adequate information on the quantitative adsorption characteristics of different tree types in relation to the radioactive pollution.

Along with the adsorption qualities of the forest, these data illustrate also a very important feature of the method, namely, the method allows us to compare the pollution on different areas. Special attention should be paid to the number of the gathered samples of the soil.

The ability of the forests to redistribute the substances in the atmosphere and the rain precipitation turned out to be high. It is clearly confirmed by the graphic representation of the distribution of the frequencies of the pollution in the forest soils (Fig.3.).

There exists a real probability (approximately 12%) to mistake that in the accidentally collected samples of the forest soil the concentration of ^{137}Cs in the neutral Taurene pine forest could be higher than in the Rucava pine forest influenced by the Chernobyl accident.

In accordance with the account the fact that the radioactive pollution gets into the soil mainly with litters of the coniferous and broadleaved trees, and on the areas with a larger amount of litter the upper layer of the soil is looser than on these areas with a thinner layer of litter, the reliability of the average data in relation to the chosen measurements Bq/kg or Bq/l could be questioned.

Our material confirms that outside the forest territories where the coefficient of the variations in soil (g/cm^3) density V is low (7-8%) the fluctuation of the quantities of radioactive ^{137}Cs is of the same kind, regardless of the use of the measurements either of volume

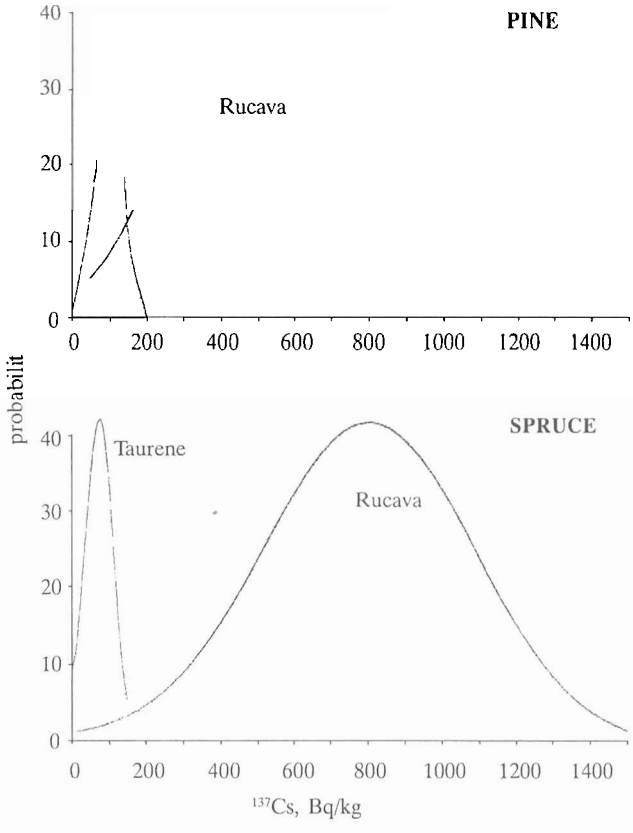

Fig. 3. Distribution of ^{137}Cs in pine and spruce forests in the influence zone of Chernobyl accident (Rucava) and in the unpolluted zone (Taurene).

or of weight: in Rucava V is 18% and 21%, in Taurene 44% and 45%. The different coefficients of variations V are in the forests. For instance, in the spruce forest in Rucava Bq/l – V=53% and Bq/kg – V=72%.

Although the average level of the pollution of the soil with ^{137}Cs in the Rucava coniferous forests is significantly higher than in the Taurene forests regardless of the measurements used, the biometrical analysis of our data certifies that using the measurement of volume Bq/l, the number of samples N in the pine forest should be not less than 10, in the spruce forest N should be more than or equal to 25, in the birch forest N>7 and outside of the forest territory N>4. By using the weight measurements Bq/kg, the number of samples should be approximately two times larger to attain the chosen 90% probability level.

Conclusions

1. The pollution of ^{137}Cs gets into the soil mainly with the litters and 95% of the pollution accumulates within the upper layer of 0-5 cm with rather different

density, that in our samples fluctuated within limits from 0,328 to 1.308 g/cm^3. The significant correlative coherence ($r = -0.63$, $r_{0.05} = 0.44$) was established between the density of the upper layer of the soil and amount of the fallen pine needles in the summer of 1995. It shows that there are places in the forest where the soil systematically is enriched with large or small amount of needles thus forming the non-homogeneous structure of the soil and pollution as well.

2. On the territories outside the forest (in Rucava) the content of ^{137}Cs in the soil is approximately three times lower than that of radiocesium in the birch forest, 20 times lower than in the pine, and 40 times lower than in the spruce forest.

3. The average indices of the radioactive pollution in the Rucava pine forest are 4.6 time higher than in the Taurene forest (446 and 97 Bq/kg); in the spruce forest this difference reaches 11.7 times (813 and 69 Bq/kg), outside the forest territory - 2.6 times (23 and 9 Bq/kg). In view of the fact that during the Chernobyl accident, the birch trees were not in leaf yet, the proportion of the soil pollution in the birch forest is only 1.5.

4. Because of the mosaic-like structure of the radioactive pollution of the soil in the pine forest 20 sampling spots are necessary; in the spruce forest 50, in the birch forest and outside of the forest territory – 10 sampling spots are needed (in our case) for obtaining statistically correct average sample (reliability level 0.90).

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ЛЕСНАЯ ПОДСТИЛКА КАК ИНДИКАТОР РАДИОАКТИВНОГО ЗАГРЯЗНЕНИЯ

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Резюме

Приведены результаты содержания ^{137}Cs в лесной подстилке в лесах Латвии после Черновыльской аварии. Исследована роль лесных насаждений в распределении и аккумуляции ^{137}Cs в почве. Проведено сравнение содержания ^{137}Cs в верхнем слое почвы сосновых, еловых, березовых лесов и на пахотных землях Руцавы (в зоне загрязнения) и Таурене (мало загрязненной зоне).

В сосновых лесах Руцавы концентрация ^{137}Cs находилась в интервале 108-724 Бк/кг, а на пахотных землях в интервале 15-30 Бк/кг.

В Таурене содержание ^{137}Cs в сосновых лесах составило 42-157 Бк/кг, еловых 19-133 Бк/кг, а на пахотных территориях 3-19 Бк/кг. Данные подтверждают большую роль хвойных лесов в повышенной абсорбации арозольных частиц, содержащих ^{137}Cs.

Ключевые слова: цезий-137, загрязнение, лесная подстилка, сосна, ель, Чернобыльская авария.