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INVESTIGATIONS ON THE RADIONUCLIDE CONTAMINATION IN THE SOILS OF SHATSK NATIONAL NATURAL PARK (THE VOLYN REGION, UKRAINE) IN THE PERIOD 1994-2000

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A b s t r a c t. The results of studies on the radionuclide contamination in the soils taken from the western part of the territory of the Shatsk National Natural Park (ShNNP, the Volyn region, Ukraine) performed in the period 1994-2000 have been presented. Basing on the experimental results, a graph of the ^{137}Cs density contamination in the soils of the investigated territory was drawn. The investigation carried out on the ^{137}Cs vertical distribution in various kinds of soils from the Park and the forecasting changes in the distribution at a depth of up to 50 cm in the case of sod loamy sandy gleyed soil of the Park for the next 75 years have been done.

K e y w o r d s: soils; radioactive contamination; radionuclides; radiocaesium; contamination density, Shack National Natural Park

INTRODUCTION

Ukraine suffered most of all countries from the Chernobyl disaster. Practically the whole of its territory was polluted by the Chernobyl fallout to some degree. A wide spectrum of radiocaesium contamination density in the soils, from insignificant up to millions becquerels per square meter, was found there. Especially, the territory of the Ukrainian Polesye, including its Volyn part, was considerably polluted. It is characterized by a wide variety of forest biocenoses with their specific soils. Many research works, including monographies ones, were dedicated to the studies of the radioactive contamination of this part of the Ukrainian territory. However, there are practically no data on the systematic in vivo monitoring of modifications of the radioactive contamination in the top soil layer over a longer period of time.

The main purpose of this work is to study the distinguishing features of the radiocaesium stock distribution in the soils of the Shatsk National Natural Park (the Volyn region, Ukraine) in the period 1994-2000 and forecast its behaviour in the upper 20 cm layer of one of the typical soils of the Park for 100 years from the initial fallout.

RESEARCH METHODS AND OBJECTS

Since 1994, we have been studying peculiarities of the radiocaesium migration in soils of ShNNP by gamma-spectrometry in the laboratory of Department of Physics, Ivan Franko National University of Lviv. The research programme allowed to determin composition of radionuclides present in the top 0-20 cm layer of the soils in the Park, density of contamination with the radionuclides of an artificial origin, and also changes in the distribution of radiocaesium stocks with depth in time, beginning from 1994. For this purpose, soil samples for the determination of contamination density were selected in 1994-1995, and then soil samples were taken annually from depth layer-by-layer up to 50 cm, with a 1 cm step, at selected location.

Soil samples for the determination of radionuclide contamination density were taken by an "envelope method" from a depth of 20 cm with a cylindrical sampler, with a cylinder bore of 40 mm. At each sampling point, five "pricks" were carried out by the sampler, then the mass selected was poured into a package. An exposure gamma dose rate was measured at each sampling point at depths of 3-4 cm and 1 m from the soil surface.

Preparation of the samples selected for gamma-analysis consisted in the purification of the mass sampled from organic and stone particles, drying, grinding in order to get grains with a diameter lower than 2 mm, and then careful mixing to homogenise. The mass prepared in this way was weighed, then part with the volume of 1 l was separated for the measurements. After weighing part of the sample was placed in a 1 l Marinelli beaker and put into the measuring chamber of the spectrometer.

The measurement of the specific activities of gamma-emitted radionuclides in the soils and contamination densities were carried out with a gamma-spectrometer, using a standard technique. The Ge(Li)-detector used had a registration of sensitivity of 120 mm^2 and a resolution (FWHM) of 2.5 keV at 1332 keV. For the reduction of the influence of the external background radiation, the detector was surrounded by a leaden protection with a thickness of 50 mm. The measuring time

was selected from the conditions to ensure the error of the specific activity of not more than 10%, with the measuring geometry used.

RESULTS AND DISCUSSION

The gamma-spectrometric analysis of the soil samples from the Park showed the presence of both a natural radionuclides (^{40}K and the representatives of uranium and thorium radioactive series such as ^{212}Pb and ^{214}Pb, ^{212}Bi and ^{214}Bi, ^{208}Tl, ^{226}Ra, ^{228}Ac, and others) and radiocaesium isotopes (^{137}Cs and much less often ^{134}Cs).

Depending on the soil type, the values of the ^{40}K specific activities determined in the soil are in the limits of 106...148 Bq kg^{-1}; the activities of the representatives of natural radioactive series did exceed 6...9 Bq kg^{-1} and, practically, did not change in time. Such a behaviour of the natural radionuclides was caused by their content in the soil formed by its geochemical structure. The above isotopes are either long-lived (e.g., ^{40}K, for which $T_{1/2}=1.2810^9$ years) or are in the radioactive equilibrium with other members of appropriate radioactive series [3].

Radiocaesium isotopes are of an artificial origin. They can get to the environment either from nuclear tests or nuclear failures, such as, the Chornobyl accident. The identification of contamination origin is possible if a cotnent ratio of these radionuclides in the soil at the moment of measurement taking is used. ^{134}Cs which as a result of entered the environment of nuclear tests is now practically completely decayed. In the Chornobyl depositions, ^{137}Cs activity at the time of failure was approximately two times higher than ^{134}Cs [5,6]. As both radionuclides are isotopes of the same chemical element, their properties in migration processes should be similar and changes in their activities in the contaminated objects in time should be only determined by the natural decay. The $^{137}Cs/^{134}Cs$ activity ratio as at January 1, 1995 determined for various samples, was equal to 343, and was in good agreement with the value of about 35 expected for the Chornobyl failure. It proved that the radiocaesium contamination of different objects in the Park is of the Chornobyl origin.

In order to determine the ^{137}Cs contamination density in the top 0-20 cm soil layer taken from the Western part of the Park territory limited by the western border of the Park and the Svityaz and Luky lakes, 254 soil samples were taken in 1994-1995 and their gamma-emitted radionuclide content was analyzed. The above results, were used for the construction of a three-dimensional plot of the ^{137}Cs contamination density distribution (Fig. 1). Distribution of the radionuclide

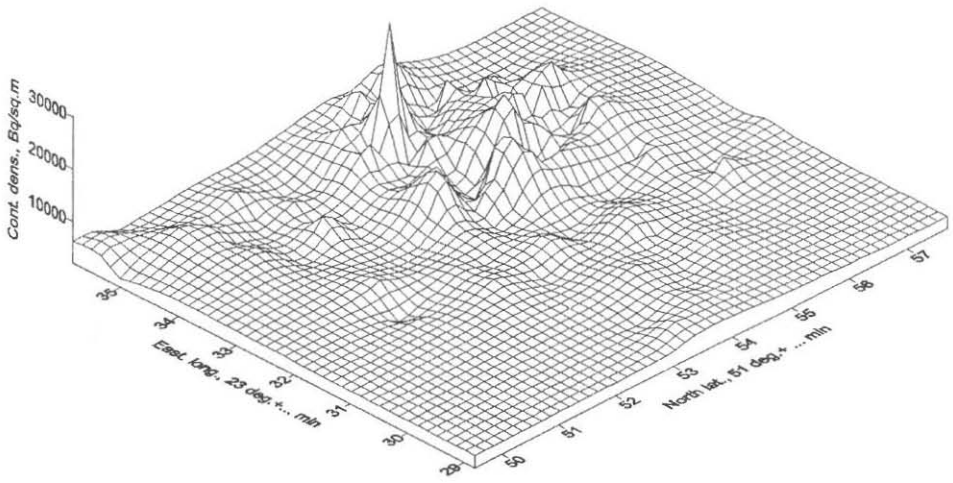

Fig. 1. The 137Cs contamination density for the soils from a northern-western part of the territory ShNNP (referred to January 1, 1996)

content in the soils taken from various spots covering a significant part of the territory investigated differ considerably and form a "mosaic structure" of contamination in the topsoil. This is especially characteristic, for the wood regions. The ^{137}Cs contamination density of the soil varies in the range of 2600 Bq m^{-2}...8000 Bq m^{-2}. Results of earlier studies [8] on the contaminated territories showed that the mosaic structure of the radioactive contamination originating from the Chernobyl failure was typical for the forest soils in the territory of the Ukrainian Polesye.

The formation of such contamination structure could be influenced by the following factors. Type of soil and migration processes occuring in the soils and causing movement of radionuclides among the soil layers, may play a significant role. As it is well known [1,11,13], radionuclide migration through the soil is determined by the phase of the initial fallout (of fuel particles or condensed components), differences in the morphological structure of the soil (in particular, the presence or absence of clay complexes), the soil acidity, as well as a hydrologic mode inherent at each point. However, it is obvious that one of the most important factors which influence formation of the mosaic structure of the topsoil pollution, is the presence of certain kinds of vegetation, in particular, coniferous trees, as their litter keeps radiocaesium for a long time [14], or else some representatives of berries, mosses, and etc.

It is known that coniferous forests acted as specific filters after the Chornobyl accident, accumulating a significant part of radioisotopes originating from the initial fallout in the pine needles of tree crowns [5]. Taking into account the fact that the replacement of pine-needles of the crown takes several years, while mineralization of the decayed pine-needles and, accordingly, transition of the radionuclides accumulated in them to the forms accessible for migration into the environment (including soils) take more than 5 years [8,14]. Then the radiocaesium from the initial Chernobyl fallout must have been kept in the coniferous dumps and the forest litter for all this significant period of time, in an inaccessible state as far as the migration processes are concerned. However, after mineralization of the coniferous dumps a significant part of radionuclides accumulated in the coniferous litter became mobile and began to arrive intensively into the soil and, accordingly, migrate in there due to various natural factors (first of all, under the influence of moisture flows). Thus, owing to the peculiarities of the radionuclides transition from litter to soil in coniferous forests, there appeared conditions for a significant delay in the transition of radionuclides into the soil depth. This may be the reason for the formation, of a higher density of radioactive contamination (when compared to open places) in the topsoil of coniferous forests in time.

One more factor that can cause differences in the content of radiocaesium in the top soil layers (or on its surface) is the presence of a moss cover. Mosses and lichens usually cover the ground with a thick continuous layer, and, consequently, after the initial fallout, radionuclides could not practically get into the soil, as they became engulfed with a living tissue of those plants and kept fixed in it for a long time [2]. Hence, radioactivity contamination density of the soil in the spots where the moss cover was present can differ from the corresponding values for the same soils without the moss.

Patches of berries are mainly characterized by the soils of peat-bog types. According to [1,10], radionuclides in those soils possess a higher ability to migrate than the typical for the other types of soils, including a soil-to-plant migration. Berries (*Vaccinium vitis-idaea L.*, *Vaccinium uliginosum L.*, and, in particular, *Vaccinium murtilus L.*) possess a significant ability to accumulate radiocaesium from the topsoil into the foliage [7,8] which is falling down later on. Thus, radionuclides in the litter are regenerated annually. Taking into account that the time needed for the foliage litter mineralization equals to one or two years [11], we can conclude that radiocaesium previously transferred from the soil to the vegetative organs of plants, will be taken up again into the processes of migration with a delay mentioned above. Thus, this specific "cycle" of radiocaesium transfer in the

soil-plant chain which ensures its constant circle from the root layer of the soil to its surface, can obviously result in differences of the radionuclide content measured in the topsoil of the patch of berries as compared to the soil content where berries are absent.

Depth distributions of the ^{137}Cs specific activities down to 35 cm for some soils in the ShNNP collected in July 1997 has been represented in Fig. 2. For a big depth (down to 50 cm), radiocaesium content did not exceed the sensitivity limit (0.5 Bq kg^{-1}). Densities of radiocaesium soil contamination at the points of sample differed notably and were as follows: 0.10 Ci km^{-2} (for sample 1); 0.09 Ci km^{-2} (2); 0.08 Ci km^{-2} (3); 0.10 Ci km^{-2} (4); 0.175 Ci km^{-2} (5).

As can be seen in Fig. 2, significant differences in the character of radiocaesium depth distribution for various types of soils in the Park were observed. For the sod-hidpodzolic (piny sandy) sandy soil (curve 1), changes in the depth distribution were almost exponential. As was mentioned above, it was peculiar of this soil type that a large part of radiocaesium stocks was concentrated in the soil litter (more than 50%, in this case) and in fresher coniferous dumps (about 15%). Moreover, practically all radiocaesium present in the soil was concentrated in the topsoil (0-5 cm). The exponent change in the depth distribution in this soil was similar to the profiles observed during the first year after the Chornobyl accident [12,14]. As was noticed in [14], radiocaesium in the soluble phase deposited in the soil from litter, was not at once fixed in the top soil layer, but penetrated deeper

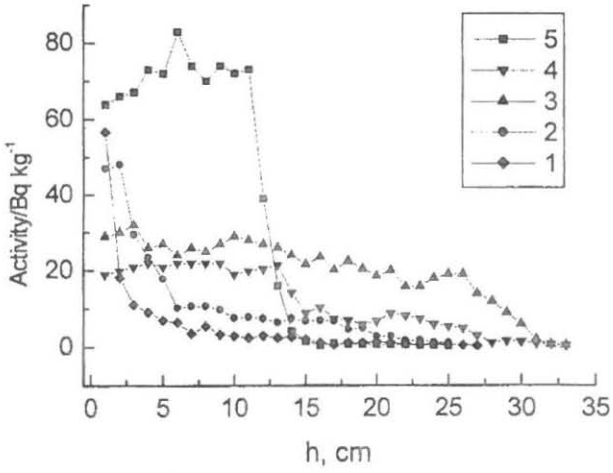

Fig. 2. The depth distribution of the ^{137}Cs specific activities in the soils of ShNNP (July 1997): 1, sod-hidpodzolic (piny sandy) sandy soil; 2, sod-weakly podzolic sandy soil; 3, sod-weakly podzolic subsandy soil; 4, sod subsandy soil; 5, sod-weakly podzolic gleyish subsandy soil

and was gradually absorbed, forming an appropriate exponential profile of the depth distribution.

The depth distribution of the ^{137}Cs activity was similar to the one observed at a depths of more than 2-3 cm in the sod-weakly podzolic sandy soil (Fig. 2, curve 2). For other soil types, the relation between the radionuclide depth distribution differed a lot from the exponential one. However, while a monotonous decrease in the radiocaesium content with depth was observed in the sod-weakly podzolic subsandy soil (curve 3), depth relations in the sod subsandy soil (curve 4) and the sod-weakly podzolic gleyish subsandy soil (curve 5) were similar: the ^{137}Cs content slowly increased down to a depth of 12-13 cm, and then decreased is almost exponential with its further depth increase in depth.

Since all samples were selected almost at the same time during the first half of July, 1997 in the territory of the Park (all sampling places were situated within a radius of 5 km from the Pisochne lake), it was possible to eliminate the influence of differences in the climatic conditions on the character of profiles of the radiocaesium contamination in the top soil layers. Accordingly, the physical-chemical phase (i.e., correlation between fuel and condensed components) at the initial fallout cannot essentially differ at the sampling points. Hence, formation of these profiles (and their temporary changing) is mainly affected by the processes related to differences in the physical-chemical soil structure and hydrological modes at the sampling points, as well as peculiarities of transition of the deposited radionuclides from the soluble to the fixed phase, and vice versa. A significant role is played by the uniformity of the soil studied layer.

Dynamics of the changes in time of the ^{137}Cs depth profiles in the upper layers of the sod-weakly podzolic gleyish subsandy soil from the territory of the Park (Fig. 3) was predicted by a dispersion migration model [1,6,8-10,13] taking into account diffusion, directed carrying towards radionuclides into the bulk of the soil and its natural decay. Calculations were carried out using the following equation [1,9]:

$$q(x,t) = Qe^{-\lambda t}\left\{\frac{1}{\sqrt{\pi Dt}}e^{-\frac{x-wt}{4Dt}} - \frac{w}{2D}e^{-\frac{wx}{D}}erfc\left(\frac{x}{2\sqrt{Dt}} + \frac{w}{t}\sqrt{\frac{t}{D}}\right)\right\}, \qquad (1)$$

where $q\ (x,\ t)$ is a relative concentration of radionuclides in the soil layer at a depth x at time t; Q – concentration of radionuclides in the boundless thin top soil layer at the initial moment; a special function.

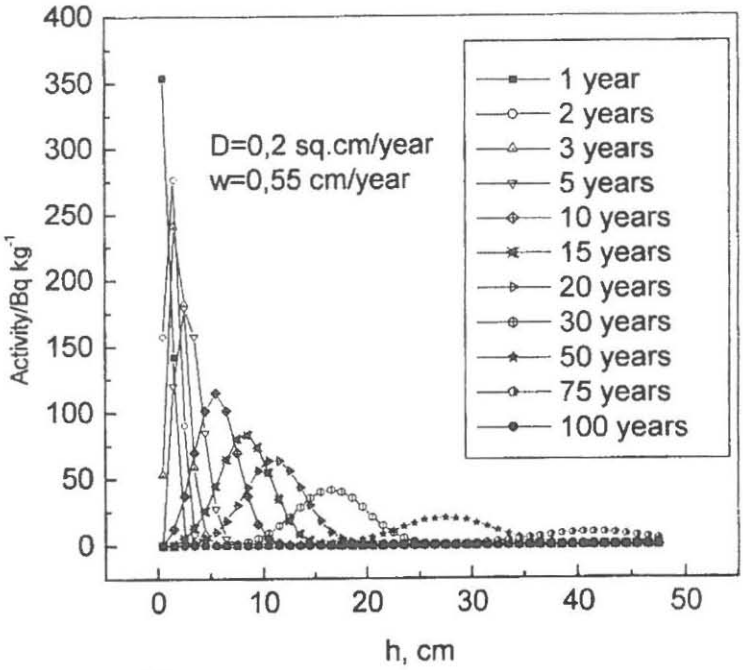

Fig. 3. A forecast for the ^{137}Cs migration in the od-weakly podzolic gleyish subsandy soil

Using a compartment model [4,6,13] and basing on the depth distributions of ^{137}Cs special activities in the soil measured in 1994-2000, appropriate factors of D diffusion and the directed transfer w were calculated for each layer. The values of the factors obtained for the whole depth profile were averaged to enable the calculating of an effective D and w values (D=0.2 $cm^2\ y^{-1}$ and w=0.55 $cm\ y^{-1}$, respectively; note that the above values were slightly higher than the estimations for the radiocaesium migration mobility in the soils of the Ukrainian Polesya [10], even though they were in agreement with other data [6]). Contamination of a film type determined according to the total ^{137}Cs stocks in the soil of the Park in July 2000 and compared to the state in May 1986 (taking natural decay into account) the initial pollution level.

It can be seen from Fig. 3 that from the most important points of view relating to the formation of an external irradiation dose and radionuclides transition from the soil into plants, radiocaesium can migrate to deeper layers due to diffusion processes and a convective transfer from the topsoil (0-20 cm), not earlier than after 50 years after the initial fallout, i.e., not earlier than in 2035.

CONCLUSION

Results of gamma-spectrometric analysis showed that in all soil samples from the Park, representatives of the uranium and thorium radioactive series, as well as ^{40}K (natural radio-nuclides) and radiocaesium isotopes (artificial radionuclides of the Chernobyl origin) were present. Basing on the results for the radionuclide contamination, a 3D plot of the ^{137}Cs density contamination of the soils was drawn for all investigated sites. The "mosaic structure" of soil contamination due to the Chernobyl fallout was noted.

It can be predicted that radionuclides transition from the soil into plants; would take place radiocaesium can migrate from the topsoil (0-20 cm) into deeper layers due to the diffusion and convective carry processes during a period not shorter than 50 years after the initial fallout, i.e., not earlier than in 2035.

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BADANIA ZANIECZYSZCZEŃ RADIONUKLIDAMI GLEB SZACKIEGO PARKU NARODOWEGO (REGION WOŁYŃSKI, UKRAINA) W LATACH 1994-2000

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S t r e s z c z e n i e. W pracy przedstawiono wyniki badań zanieczyszczeń radionuklidami próbek gleb skażonych wybuchem elektrowni w Czernobylu, pobieranych w latach 1994-2000 z zachodniej części terenu Szackiego Parku Narodowego. Stwierdzono zawartość w glebach zarówno naturalnych radionuklidów, jak również radioaktywnego ^{137}Cs i w mniejszej części ^{134}Cs. Przewidziano, że zmiany stężenia cezu w warstwie 50 cm gleby będą następować przez następne 75 lat.

S ł o w a k l u c z o w e: gleby, skażenie radioaktywne, Szacki Park Narodowy, Ukraina