

Impact of sharp weather warming on the exchangeable forms of ^{137}Cs in soils and its bioaccumulation in orchard grass

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Accepted: August 28, 2024

The manuscript considers the effects of the sharp temperature increase on the exchangeable forms of ^{137}Cs , from one to three months after its entrance in the soil. The impact of the sharp increase of the environmental temperature on the migration ability of ^{137}Cs and bioaccumulation in orchard grass was investigated in soils taken from five different regions in Bulgaria. The soil samples were contaminated with an aqueous solution of ^{137}Cs and stored at different temperature regimes, simulating temperate and hot summer conditions for a period from one to three months. The exchangeable forms of the radionuclide were determined by single extractions with 1 M NH_4NO_3 , followed by gamma-spectrometric determination. Vegetation experiments with the conditioned soil samples were carried out and the transfer factors (TFs) of orchard grass (*Dactylis glomerata* L.) were determined. The results showed that the conditioning for one month at increased temperature, two months after contamination caused a decrease in the exchangeable ^{137}Cs and the transfer factors. A reduction of the exchangeable forms of radiocesium with the increase of the content of muscovite in the soils was observed.

It was found that the change in TFs provoked by the temperature raise two months after contamination increased with increasing potassium concentration in the soils.

Key words: ^{137}Cs ; soils; sharp warming; exchangeable forms; transfer factors; orchard grass

INTRODUCTION

Studies of the migration and geochemical forms of artificial radionuclides in the environment became particularly intensive in the 20th century and were provoked by nuclear experiments and nuclear power plant accidents which resulted in radioactive isotopes entering the air, water and soil in dangerously high activity levels. After being released into nature, radioactive isotopes are distributed into the ecosystem components through different pathways, reaching the human body most often through food chains. Many radionuclides are retained in the soil by adsorption on the clay mineral particles, by ion exchange, precipitation as hydroxides or sulfides, or by complexation with humic substances. The geochemical forms of radionuclides in soils and their transfer to plants have been intensively studied by different research groups [1-4] in order to determine the risk of their spread in nature and within the food chain and to select an appropriate method of remediation [5]. The migratory ability and bioavailability of radionuclides in soil are determined by a number of factors, such as the soil characteristics (soil pH, texture, cation exchange capacity, organic matter content, etc.), climatic conditions, addition of fertilizers, plant species and land use [6-8]. The

International Atomic Energy Agency (IAEA) has prepared detailed reviews of radionuclide transfer factors for various food products [9, 10], based on the results of numerous studies carried out, which allow to predict the degree of accumulation of pollutants in the event of a nuclear accident.

In the recent decades, abrupt variations in climatic parameters have been recorded, causing changes in soil characteristics and thus affecting the fate of pollutants in nature. This requires the need to obtain more data on the mobility of radioactive pollutants in natural systems under conditions of sudden warming or cooling, in the events of drought, or flooding. Thus, the geochemical behavior of contaminants could be better understood and measures taken against their spread through the food chain. Our previous investigations showed that the storage of radioactively contaminated soils at different temperatures and soil moistures can lead to a change in radionuclides geochemical forms and thus affect their transfer to plants. These effects were dependent on the soil characteristics and radionuclide chemical properties [11-14].

^{137}Cs is one of the long-lived technogenic radionuclides that is very important from the point of view of radiological protection. It is β and γ emitter, with a half-life of 30.2 years and is produced

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as a fission-product in nuclear reactors and aboveground nuclear weapon explosions [15]. Cesium has low mobility in the environment and is mainly fixed in soil. The concentration bound in the sandy soil particles was estimated to be 280 times greater than in the water contained in the pores between the soil particles [16]. Interest in the levels of radiocesium contamination in soils and its transfer along the food chain has raised, especially after the Chernobyl accident in 1986 and later in Fukushima in 2011, as a result of which the maximum permissible levels of radiocesium in various food products after accidents have been set [17-19]. The knowledge of soil-to-plant transfer factors or the possibility of their prediction under given conditions is a significant advantage when preparing risk assessments in case of radioactive contamination. A number of studies carried out earlier showed the relationship between the concentration of potassium in the soil and the transfer of radiocesium from the soil to the vegetation [1, 20-22]. Therefore, the complex influence of mineralogical composition, soil characteristics, potassium concentration, and environmental conditions (such as temperature and soil moisture) is worth investigating to gain insight into the possible effects of the abrupt warming on radiocesium migration and transfer.

The aim of this paper is to present the results of a study on the effect of abrupt weather warming on the exchangeable forms of ^{137}Cs in soils, as well as on its bioaccumulation in orchard grass. The study is a model experiment simulating soil contamination with an aqueous solution containing $^{137}\text{Cs}^+$ and conditioning at 18 °C and 40 °C for a period from one to three months. The effects of the different temperature regimes on the exchangeable forms of radiocesium and on its transfer factors are presented, taking into account the composition of soil minerals and the concentration of potassium, as its main competitive ion in the soil to plant uptake.

MATERIALS AND METHODS

Soil samples and conditioning

For the purposes of the present study, soils with different soil characteristics were taken. Soil samples were taken from the surface 0-10 cm layer. The samples were air-dried, cleaned of mechanical impurities and sifted through 2 mm hole size sieves.

The soils were taken from five different regions in Bulgaria and classified according to World Reference Base for Soil Resources/FAO [23] as follows: Albic cambisol, Calcaric chernozem soils, Gleyic fluvisol, Salic fluvisol and Vertisol. Weight of 500 g of each soil type was contaminated with a solution of ^{137}Cs in nitrate form. The soil samples

were homogenized and watered, maintaining soil moisture of 20-25 wt% in open-air, at 18 °C for two days. Afterwards each soil sample was divided into subsamples and each of them was conditioned at the following regimes:

Regime A1: One month at 18 °C and soil moisture of 20-25 wt%.

Regime A2: One month at 40 ± 0.1 °C and relative air humidity of 50 ± 0.1 wt % achieved by using a climate chamber Model HPP 108 (Memmert GmbH, Germany). The samples were watered every two days and the soil moisture varied within the range 5–50 wt %, causing cycles of drying and wetting.

Regime B1: Two months conditioning at 18 °C and soil moisture of 20-25 wt%.

Regime B2: One month at 18 °C and soil moisture of 20-25 wt%, followed by one month at 40 ± 0.1 °C, relative air humidity of 50 ± 0.1 wt % and soil moisture 5–50 wt %, causing cycles of drying and wetting.

Regime C1: Three months conditioning at 18 °C and soil moisture of 20-25 wt%.

Regime C2: Two months conditioning at 18 °C and soil moisture of 20-25 wt% followed by one month at 40 ± 0.1 °C, relative air humidity of 50 ± 0.1 wt % and soil moisture 5–50 wt %, causing cycles of drying and wetting.

The chosen temperature regimes aimed to throw light on the influence of rapid warming over a period of one month on the exchangeable forms of ^{137}Cs one, two and three months after soil contamination. Aliquots of each soil were taken after the end of each conditioning period, and the exchangeable forms of radiocesium were determined.

Vegetation experiment

After that, the conditioned soils were placed at a temperature of 18 °C and planted with orchard grass (*Dactylis glomerata* L.). The grass grew for two weeks at temperate conditions; grass samples were collected by cutting at 0.5 cm above the soil surface, washed with tap and distilled water, air-dried, grinded in a ball-mill and the specific activities of ^{137}Cs were measured using gamma-spectrometry.

Soil characterization

The main characteristics of the selected soils: pH in KCl and in H_2O (soil: water ratio 1:10), humus and the size distribution of soil particles were determined in order to identify the soil texture.

The humus content was determined by the method of Tjurin [24].

Cation exchange capacity (CEC) was determined after soil extraction with 1 M $\text{NH}_4\text{CH}_3\text{COO}$ (Trace Select, Sigma Aldrich) and measurement of the

concentrations of Ca²⁺, K⁺, Mg²⁺ and Na⁺ in the supernatant by ICP-MS (Perkin Elmer DRCE).

Determination of potassium concentration in the soil samples was performed after microwave digestion and analysis by ICP-MS (Perkin Elmer DRCE) according to the procedures described in [25]. Five standard reference materials: reference stream sediments STD-1, STD-3 and STD-4 (Canada Center for Mineral and Energy Technology, Geological Survey of Canada), IAEA were used for estimation of the accuracy of the method. The analytical recoveries determined valued from 99.3 to 99.8 %.

The crystal structure of the selected soils was studied by X-ray diffraction method (powder diffractometer Siemens D500) using CuKα radiation filtered by a secondary monochromator (40 kV, 30 mA, 0.05_ 2H/2 s) for the 2H interval 3–60_ . The phase identification and quantitative phase analysis of the soil minerals were performed by X'Pert HighScore Plus software.

Extraction

The exchangeable forms of ¹³⁷Cs were determined by single extractions with 1 M NH₄NO₃ for 1 h, followed by gamma-spectrometric determination. Detailed description of the procedures was given in [26].

Gamma-spectrometry

HPGe detector Canberra 7221 coupled to a 16000-channel analyzer DSA-1000 was used for the detection of ¹³⁷Cs. The gamma spectra were processed using Genie-2000 Basic Spectroscopy Software. The activities of ¹³⁷Cs in the samples were determined using the gamma peak of ^{137m}Ba (daughter product of ¹³⁷Cs) at 661.5 keV, 25 min

after the sample preparation in order to reach radioactive equilibrium.

Calculations

To determine the transfer of radiocesium from the soil to the plant, the transfer factors (TFs) were calculated using the ratio:

$$TF = \frac{\text{Specific radioactivity in grass [Bq/g]}}{\text{Specific radioactivity in soil [Bq/g]}} \quad (1)$$

The effects of rapid warming applied during the regimes A2, B2 and C2 were assessed by calculating the relative changes (Δ, %) of the exchangeable forms and TFs as compared to regimes A1, B1 and C1, using the following formulas:

$$\Delta_{\text{exch}\%} = \frac{C_{\text{sexch}}(40^\circ\text{C})\% - C_{\text{sexch}}(18^\circ\text{C})\%}{C_{\text{sexch}}(18^\circ\text{C})\%} \times 100 \quad (2)$$

$$\Delta TF\% = \frac{TF(40^\circ\text{C})\% - TF(18^\circ\text{C})\%}{TF(18^\circ\text{C})\%} \times 100 \quad (3)$$

where % ¹³⁷Cs_{exch} is the % amount of ¹³⁷Cs in exchangeable form and TF is the transfer factor of ¹³⁷Cs after conditioning at the given temperature regime.

RESULTS AND DISCUSSION

The main characteristics, the mineral composition of the soil crystal phases and the concentrations of K are presented in Table 1.

The results from the exchangeable forms of radiocesium after conditioning of the soils at the different temperature regimes A1 - C2 are established on Fig. 1. Mean values from measurements of three parallel samples are shown.

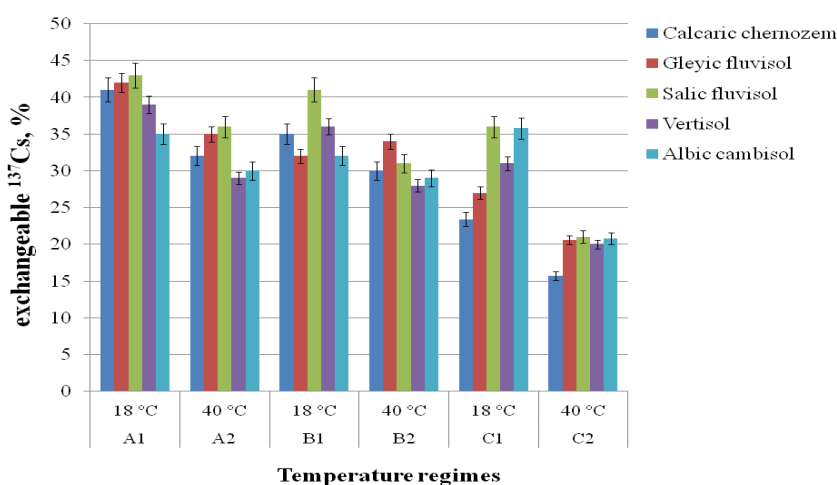


Fig. 1. Exchangeable forms of ¹³⁷Cs (%) in the studied soils, after conditioning at the different temperature regimes A1 – C2.

Table 1. Main characteristics of the investigated soils: mineral composition and potassium concentrations [mg/kg] and standard uncertainties

| Soil order/ Texture class | S o i l p H | | CEC (meq/100 g) | Humus, % | Mineral composition, % | K [mg/kg] |
|----------------------------------|------------------|-----|--------------------|----------|--|-----------|
| | H ₂ O | KCl | | | | |
| Calcaric chernozem/ Silt loam | 7.8 | 7.4 | 13.2 | 3.18 | Vermiculite Mg,Fe,Al) ₃ (Al,Si) ₄ O ₁₀ (OH) ₂ x ₄ (H ₂ O) -0.2 % Muscovite KAl ₃ Si ₃ O ₁₀ (OH) ₂ -3.9 % Quartz SiO ₂ -39.4 % Albite NaAlSi ₃ O ₈ -5.0 % Magnesium calcite CaMg(CO ₃) ₂ - 2.0 % Pyroxene Li _{0.23} Mg _{1.54} O ₆ Sc _{0.23} Si ₂ - 49.6 % | 1977 ± 49 |
| Gleyic fluvisol/ Loamy sand | 7.9 | 7.7 | 9.80 | 3.09 | Vermiculite Mg,Fe,Al) ₃ (Al,Si) ₄ O ₁₀ (OH) ₂ x ₄ (H ₂ O) - 0.5 % Muscovite KAl ₃ Si ₃ O ₁₀ (OH) ₂ - 0.7 % Quartz SiO ₂ - 41.4 % Albite NaAlSi ₃ O ₈ - 8.4 % Magnesium calcite CaMg(CO ₃) ₂ - 2.0 % Pyroxene Li _{0.23} Mg _{1.54} O ₆ Sc _{0.23} Si ₂ - 47.1 % | 921 ± 8.4 |
| Salic fluvisol/ Loamy sand | 5.5 | 5.2 | 5.9 | 2.65 | Muscovite KAl ₃ Si ₃ O ₁₀ (OH) ₂ - 10.4 % Diopside CaMgSi ₂ O ₆ - 7.1 % Quartz SiO ₂ - 25.4 % Paracelsian BaAl ₂ Si ₂ O ₈ - 0.8 % Feldspar KAlSi ₃ O ₈ - 56.3 % | 635 ± 9.3 |
| Vertisol/ Sandy loam | 7.3 | 7.0 | 12.80 | 4.95 | Muscovite KAl ₃ Si ₃ O ₁₀ (OH) ₂ - 6.4 % Quartz SiO ₂ - 33.4 % Albite NaAlSi ₃ O ₈ - 7.9 % Magnesium calcite CaMg(CO ₃) ₂ - 3.9 % Sodalite Na ₈ Al ₆ Si ₆ O ₂₄ Cl ₂ - 11.9 % Pyroxene Li _{0.23} Mg _{1.54} O ₆ Sc _{0.23} Si ₂ - 36.5 % | 1183 ± 24 |
| Albic cambisol/ Sandy loam | 7.6 | 7.4 | 31.10 | 2.03 | Vermiculite (Mg,Fe,Al) ₃ (Al,Si) ₄ O ₁₀ (OH) ₂ x ₄ (H ₂ O) - 0.5 % Muscovite KAl ₃ Si ₃ O ₁₀ (OH) ₂ - 17.3 % Quartz - SiO) - 69.8 % Albite NaAlSi ₃ O ₈ - 12.4 % | 1496 ± 71 |

The obtained results showed that the exchangeable forms of ¹³⁷Cs after the first month of contamination varied in the interval from 35% to 43%. The lowest values of the mobile forms are registered for the Albic cambisol soil which is characterized by the highest CEC and the lowest humus content. The highest % exchangeable forms of Cs are found in the Salic fluvisol soil, which has the lowest CEC value. This agrees with literature data, according to which CEC has a dominating role in controlling Cs mobility [27, 20] and is explained by the fact that a higher CEC indicates a greater ability of the soil to bind positive ions. In addition, the soil organic matter often covers the surface of the soil particles, which impedes the interaction between the metal ions and the inorganic minerals. Thus, the

lowest humus content in Albic cambisol contributes to the lowest exchangeable forms of the radionuclide. Cs is sorbed by ion exchange into interlayer sites of the clay particles, and by surface complexation with hydroxy groups constituting broken bonds on edge sites, and the planar surfaces of oxide and silicate minerals [27]. It was also noticed from Table 1 that the content of muscovite (in %) in Albic cambisol soil has the highest values among the studied soils - 17.3 %. The micaceous minerals, such as muscovite were found to preferentially sorb and hold trace quantities of radiocesium after entering the soils and waters [20, 28, 29]. The sorption sites for Cs of these minerals are divided into two types: the Regular Exchange Complex (REC) sites which are mainly planar, and

the Frayed Edge Sites (FESs) which are located at the edge and/or interlayer of the micaceous clay minerals [29]. The interaction of Cs with the REC is considered to be weak but FESs have a very strong affinity for this radionuclide.

The relative change ($\Delta_{\text{exch}}\%$) of the exchangeable forms after rapid warming, achieved by regimes A2, B2 and C2, as compared to the conditioning at temperate regimes A1, B1 and C1 is plotted as a function of the content of muscovite (%) and presented on Fig. 2.

Clearly pronounced decrease of the exchangeable forms of ^{137}Cs after conditioning at higher temperature for a period of one month was observed. This effect was noticed one, two and three months after contamination. Only in Gleyic fluvisol soil a slight increase of 7% was registered. The effect of decrease of exchangeable ^{137}Cs can be explained by the mechanical weathering provoked by sharp changes of the temperature and soil moisture (regimes A2, B2 and C2) which might be leading to FESs formation on micaceous minerals, thus supporting the selective retention of radiocesium from the soil solution. The drying-wetting cycles, simulated during regimes A2, B2 and C2, might have accelerated the dehydration of the clay interlayers, thus leading to subsequent interlayer collapse which might have caused the Cs trapping. Such effect of the drying-wetting cycles on the immobilization of Cs was reported in [30].

Fig. 2 shows a lack of dependence of the change of the exchangeable forms of ^{137}Cs on the muscovite content, after conditioning at regimes A2 and B2. Clearly pronounced decrease in exchangeable forms

with the increase of % muscovite in the soils is seen after storage under C2 regime, i.e. three months after contamination. This can be due to the slow diffusion of Cs^+ ions from the soil solution through the mica surfaces to the selective sorption sites. Kwong-Moses *et al.* [28] investigated the sorption and desorption of radiocesium by muscovite for a period from 18 h to 130 days. Cs was found to slowly reach the highly Cs-selective sites of muscovite, which are in the interlayer wedges formed by splaying of aluminosilicate layers during weathering of muscovite [28]. Apparently, a sharp warming for one month, two months after radioactive contamination, can lead to a reduction in the mobile forms of ^{137}Cs . This effect increases with the increase of the content of muscovite in the soils. Conducting studies with a larger number of soil orders would enrich the knowledge about the relationship between soil characteristics and the recorded effects of the sudden warming. This would enable predictive modeling of the studied systems to be used in risk assessments. Fig. 3 presents the TFs of radiocesium from the studied soils to orchard grass after conditioning of regimes A1 – C2. The TF values vary from 1.10^{-2} to $8.5.10^{-2}$, which is in agreement with our previous studies [14] and fits within the variation range of TF for ^{137}Cs from soil to grass, published by the IAEA for all types of soil as follows: mean 6.3×10^{-2} , min 4.8×10^{-3} , max 9.9×10^{-1} [10]. Decrease of the TF values after sharp temperature increase for one month is noticed for all soils and regimes. This corresponds to the decrease of the exchangeable forms of radiocesium as a result of sudden warming.

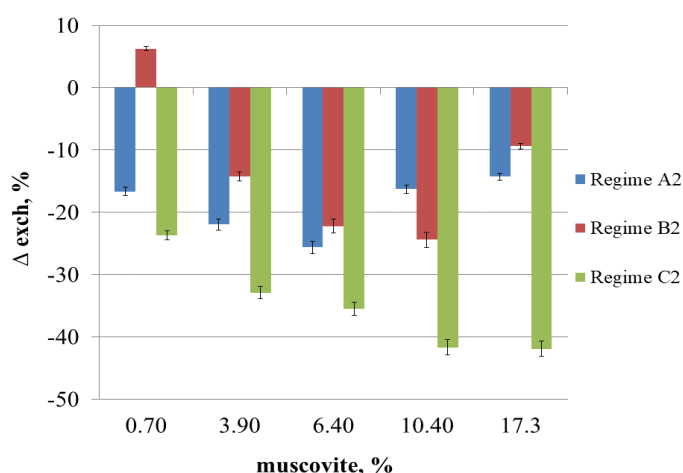


Fig. 2. Dependence of the change of the exchangeable forms of ^{137}Cs ($\Delta_{\text{exch}}\%$) on % content of muscovite ($(\text{KAl}_3\text{Si}_3\text{O}_{10}(\text{OH})_2$) in the soils after the applied temperature regimes.

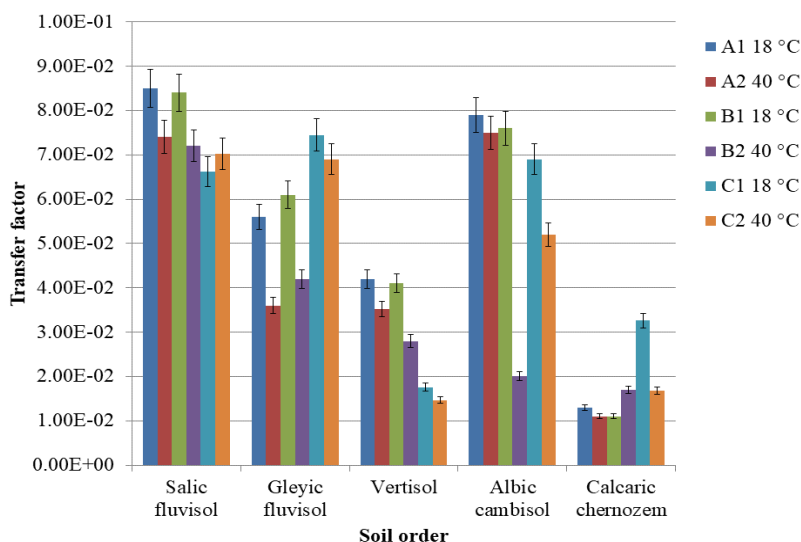


Fig. 3. Transfer factors of ^{137}Cs from the studied soils to orchard grass after conditioning at the different temperature regimes A1 – C2.

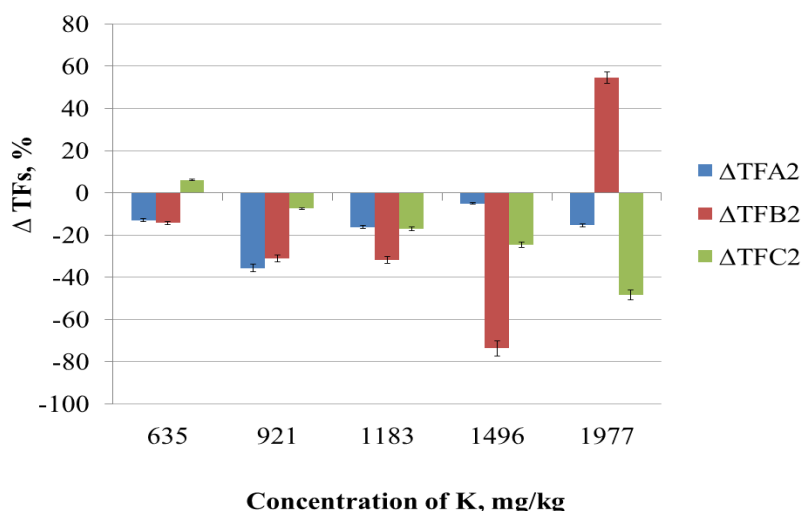


Fig. 4. Dependence of the change (ΔTF %) of the TFs as a result of sharp warming during 1 month after conditioning of the soils at temperature regimes A2, B2 and C2 on the total concentration of K in the soils.

The dependence of the change of TFs after sharp warming on the potassium concentration as the main competing ion of cesium was investigated. The results are presented in Fig. 4. It can be noticed that the changes of the TFs after A2 and B2 temperature regimes do not depend on potassium concentration in the soils. Despite the well-known competitive geochemical behavior of potassium and cesium in the uptake by vegetation [31, 32], it should be mentioned that the dynamic fixation processes during the first weeks after contamination are causing changes in the rate of bioaccumulation of radiocesium to a different degree. However, TFs obtained after storage under C2 mode showed a dependence of decreasing bioaccumulation of radiocesium with increasing total soil potassium concentration. This can be explained by the

competitive uptake of potassium from the soil solution, being in a much higher concentration, together with the selective capture of trace amounts of Cs in micaceous minerals during the period of sharp warming.

The present results contribute to the identification of soil characteristics that influence the migration ability and bioavailability of Cs under the conditions of a sharp increase in temperature. Obtaining data from a larger number of soils would allow to develop a model for prediction of the bioaccumulation of radiocesium after a period of rapid warming and drying.

CONCLUSION

The results of the conducted study of five orders of soils showed that the sudden warming for a period of one month changed the exchangeable forms of

radiocesium depending on the soil characteristics and the time that has passed since the radioactive contamination. A decrease in the exchangeable forms of ¹³⁷Cs was recorded in almost all soil samples. Clearly expressed dependence of the decrease of the exchangeable forms with an increase in the content of muscovite in the soils was established. The sudden warming was found to reduce the bioaccumulation of ¹³⁷Cs by orchard grass in most of the soil samples. A dependence of the decrease of the transfer factors soil-to-orchard grass and the total concentration of potassium in the soils after sudden warming for one month, two months after the radioactive contamination was noticed.

Obtaining such data in more soil orders and on other radionuclides would be helpful when predicting the radioecological risk in case of nuclear accidents during sharp temperature changes for different periods of time.

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