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IMPACT OF RAPID WARMING ON THE TRANSFER OF ^{60}Co, ^{137}Cs AND ^{54}Mn FROM SOIL TO GRASS AND WITHIN THE FOOD CHAIN*

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Abstract. *Rapid changes of the environmental temperature can alter soil characteristics and influence the migration ability and bioavailability of the radionuclides. Elucidation of the effects of extreme weather conditions on the transfer factors of radionuclides in different soil types is especially important for adequate risk assessment after radioactive contamination. This paper presents the impact of a rapid increase of environmental temperature for a period of one month on the bioaccumulation of* ^{60}*Co,* ^{137}*Cs and* ^{54}*Mn from three soil types to orchard grass. The experiment was performed by soil samples, taken from the surface soil layer 0-10 cm of Albic cambisol, Calcaric chernozem and Gleyic fluvisol soils (classified according to World Reference Base for Soil Resources/FAO) from Bulgaria. The samples were contaminated by a radioactive solution of* ^{60}*Co,* ^{137}*Cs and* ^{54}*Mn, separated into two subsamples and stored during one month at two temperature regimes: 15 °C and 40 °C by using a climate chamber. Afterwards the soils were planted with orchard grass and stored at 15 °C during two weeks until growing and the transfer factors were determined. The results showed that rapid warming during one month after radioactive contamination caused a decrease of the transfer of* ^{137}*Cs from all studied soils to orchard grass. The decrease of the transfer factors of* ^{60}*Co and* ^{54}*Mn from the soil with high cation-exchange capacity, higher quartz and muscovite content was determined, while the increase of the transfer factors of* ^{60}*Co and* ^{54}*Mn from the soil with very low cation-exchange capacity and lower content of quartz and micaceous minerals was registered. Prognostic maximum specific activities of the radionuclides in the investigated soils, at which milk and meat are harmless to be consumed, were calculated referring to the obtained data.*

Key words: *Radionuclides, soils, temperature increase, orchard grass, transfer factors, prognostic risk assessment*

1. INTRODUCTION

Sharp variations of environmental temperature can cause changes of main soil characteristics, such as pH, cation exchange capacity (CEC), redox potential, organic matter decomposition, which alter microbial activity, element leaching or ion-exchange processes. This might provoke changes of the geochemical forms of radioactive contaminants and influence their transfer within the food chain [1].

The effects of substantial variations of main climatic parameters can be expected to be higher in the first months after contamination, when the radionuclides are not strongly fixed to the soil phases. The impact of some extreme environmental conditions on the geochemical forms of radionuclides in non-equilibrium systems, shortly after their release into the soil, was studied and described in [2]-[5].

Assessment of the risk of distribution of radionuclides from contaminated soils to the living organisms requires knowledge on their biological accumulation to vegetation and evaluation of their subsequent uptake to the animals and humans. Soil to plant radionuclide transfer is assessed by measuring the soil to plant transfer factor (TF) or concentration factor and is defined as the ratio of the radionuclide content in the plant (or in part of the plant) to that in the soil (Bq kg^{-1} dry weight plant tissue/Bq kg^{-1} dry weight soil). The TF values of the radionuclides vary significantly and were found to depend on the soil characteristics, plant species and agricultural practice. The influence of weather variations during the growing season was also reported to be important [6]-[7].

Our previous study on Fluvisol soil, three years after its contamination, showed that the conditioning at 40 °C before the growing season caused increased bioaccumulation ^{241}Am, ^{60}Co, ^{137}Cs in orchard grass, compared to the bioaccumulation, determined after storage at temperate conditions. This effect was higher for ^{137}Cs than for ^{241}Am and ^{60}Co [8]. Additional studies on the effects of sharp temperature change on bioaccumulation of radionuclides from different soil types are needed for better assessment of the risk of their transfer in the food chain.

Transfer coefficients have been introduced to evaluate the uptake of radioactive contaminants to animals. They are widely adopted for quantifying radionuclide transfer to both milk (F_m, d L^{-1} or d kg^{-1}) and meat (F_f, d kg^{-1}) as the equilibrium ratio of the radionuclide activity concentration in milk/meat to the daily dietary radionuclide intake [7]. The knowledge of TF, F_m and F_f values for certain plant and animal species allows a predictive risk assessment of radionuclide transfer within the food chain to be performed after measuring specific activity of the contaminated soil. For this purpose, the soil type and the quantity and composition of animal's diet are to be taken into account. To assess radionuclide uptake from contaminated meat/milk to humans, a food processing retention factor, F_r, has been introduced. It is defined by

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the fraction of radionuclide activity that is retained in food after processing (e. g. frying, boiling, baking etc.) [7].

Thus, the decrease of radionuclide uptake as a result of a certain way of cooking, might be evaluated when predicting the risk of radioactive contamination to human health. Knowing the impact of sharp temperature variations on TF values for a given radionuclide, soil type and vegetation, and the availability of data for F_m, F_f and F_r coefficients, would enable to evaluate the prognostic limit of specific activity of the radionuclide in the soil, above which the corresponding food products cannot be consumed.

This manuscript presents the effects of rapid temperature increase (simulating hot summer) during one month on the transfer factors of ^{60}Co, ^{137}Cs and ^{54}Mn from Albic cambisol, Calcaric chernozem, and Gleyic fluvisol soils (classified according to World Reference Base for Soil Resources/FAO) [9] to orchard grass. The experiment modeled contamination of soils with an aqueous radioactive solution at temperate conditions (12 – 15 °C and soil moisture from 22 to 24 wt %). One week later, a sharp increase of the environmental temperature (up to 40 °C), continuing for one month, was simulated. Afterwards, the initial temperature range was restored and the soil samples were planted with orchard grass (*Dactylis glomerata L.*) for the determination of the TF values of ^{60}Co, ^{137}Cs and ^{54}Mn.

Dactylis glomerata L. was chosen for this study due to its fast growing and wide use for hay and pasture. The obtained data can be used to perform a prognostic risk assessment of radionuclide transfer from the studied soil types to the food chain at sharp temperature increase. Prognostic maximum specific activities of ^{60}Co, ^{137}Cs and ^{54}Mn in the soils, above which cow, beef and goat meat and the milk are harmful to be consumed, were calculated by using data for F_m and F_f published by the IAEA [7] and regarding maximum permissible levels of radioactivity in foods [10].

2. MATERIALS AND METHODS

2.1. Soils, contamination, conditioning and green house experiment

Albic cambisol, Calcaric fluvisol, and Gleyic chernozem soils with the total weight of 1 kg each were taken from the surface soil layer (0–10 cm) from Bulgaria. The soils were air-dried, cleaned from plant impurities and sieved through 2 mm-sieves. The general characteristics of the studied soils, including pH in H_2O and in 0.1 M KCl, CEC (cmol$^+$ /kg), relative content of sand, silt, clay, humus were determined. The mineral composition of the crystal phase of the soil samples was investigated by X-ray diffraction. Fifty-gram sub-sample of each soil was homogenized in an agate mortar and measured by gamma-spectrometry.

The specific radioactivity (S) of the radionuclides in the soils investigated prior to laboratory contamination was S < 0.6 Bq g^{-1} for ^{60}Co and ^{54}Mn; S < 0.5 Bq g^{-1} for ^{137}Cs. The rest of the bulk samples were contaminated with aqueous solutions of ^{60}Co, ^{137}Cs and ^{54}Mn in chloride forms. The specific activity of the contaminated samples was S = 20 ± 0.01 Bq g^{-1} for ^{60}Co; and S = 25 ± 0.01 Bq g^{-1} for ^{137}Cs and S = 14 ± 0.01 Bq g^{-1} for ^{54}Mn.

The contaminated samples were homogenised and conditioned for one week at 15 ± 1 °C in open-air vessels in the laboratory. The soils were watered so that the soil moistures were maintained between 20–24 wt%. Afterwards, each contaminated soil was divided into two subsamples, placed in plastic vessels, arranged in layers with thickness of 5 cm and conditioned for one month under the following conditions:

- Temperate: 15 ± 1 °C in open-air vessels in the laboratory. The soil moisture was maintained within the range of 22–24 wt %;

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

The experiments were carried out using three parallel samples of each storage condition. After conditioning, the soils were kept at temperate conditions for five days, and then planted with orchard grass (*Dactylis glomerata L.*). The orchard grass grew for two weeks at temperate conditions. The 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 prepared for gamma-spectrometric measurements by packing in standardized geometry with a weight of 15 g.

2.2. Measurement conditions

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

The radioactivity of solid samples and leachates was determined using an HPGe detector Canberra 7221 (energy resolution of 1.9 and efficiency of 16 % at 1332.5 keV) coupled to a 16000-channel analyser DSA-1000. The spectra were processed using Genie-2000 Basic Spectroscopy software. Efficiency calibration and measurements were performed as described in [4].

2.3. Calculations

Transfer factors of the radionuclides from the studied soil types to orchard grass were calculated by the ratio:

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

Prognostic maximum specific activity [Bq/kg] of the soils, above which the radioactive contamination exceeds the established permissible limits, was calculated as follows:

$$A[Bq/kg]_{\max} = \frac{A[Bq/kg]_{\text{limit}}}{F[d.L^{-1}, kg^{-1}]_{m,f}.m[kg]_{grass}.TF} \quad (2),$$

where A[Bq/kg]$_{\text{limit}}$ – maximum permissible specific activity of given radionuclide in a food product; F$_{f,m}$[d.L^{-1},kg^{-1}] – radionuclide transfer coefficient to meat, or milk of given animal species; m[kg]$_{\text{grass}}$ –dry weight of grass, taken daily by given animal species; TF – determined transfer factor of given radionuclide from soil to orchard grass, after storage at given temperature conditions.

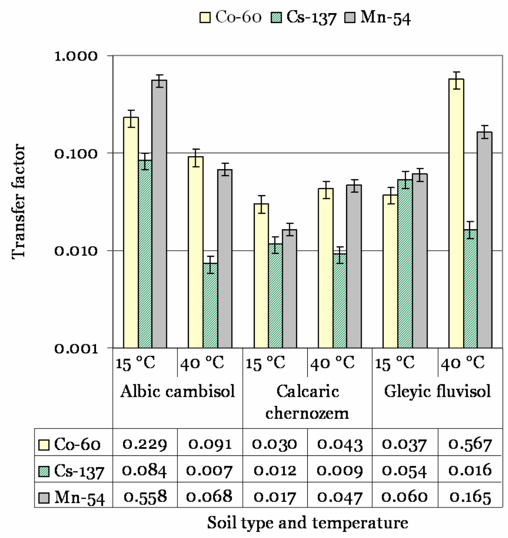
3. RESULTS AND DISCUSSION

3.1. Transfer factors

The general characteristics of the investigated soils are presented on Table 1. The studied soil types have different textures, CEC and soil mineral contents. Albic cambisol has a normal range of CEC (from 15 to 40 cmol$^+$/kg), sandy loam texture, lowest content of humus and highest content of quartz and muscovite, as compared to the other two soils. Calcaric chernozem belongs to the soils with slightly low CEC (from 10 to 15 cmol$^+$/kg), has silt loam texture, highest content of pyroxene and much lower content of muscovite. Gleyic fluvisol soil has very low CEC (from 0 to 10 cmol$^+$/kg), loamy sand texture, alkaline pH and the lowest content of micaceous minerals.

Table 1. General characterization of the investigated soils

Soil type **Characteristic**	**Albic cambisol**	**Calcaric chernozem**	**Gleyic fluvisol**
Sand, %	61.6	24.71	77.73
Silt, %	33.2	70.18	19.13
Clay, %	5.2	5.11	3.14
Texture class	Sandy loam	Silt loam	Loamy sand
Humus, %	2.03	3.18	3.19
CEC (cmol+/kg)	31.1	13.16	9.80
pH (H_2O)	7.4	7.4	7.7
pH (KCl)	7.6	7.8	7.9
Soil mineral content (%)	Quartz (70 %); Muscovite (17.3 %); Albite (12.2 %); Vermiculite (0.5 %)	Pyroxene (49.6%), Quartz (39.4%), Albite (5%), Muscovite (3.9%), Magnesium calcite (2%), Vermiculite (0.2%)	Pyroxene (47.1%), Quartz (41.4%), Albite (8.4%), Magnesium calcite (2%), Muscovite (0.7%), Vermiculite (0.5 %)



	Albic cambisol 15 °C	Albic cambisol 40 °C	Calcaric chernozem 15 °C	Calcaric chernozem 40 °C	Gleyic fluvisol 15 °C	Gleyic fluvisol 40 °C
Co-60	0.229	0.091	0.030	0.043	0.037	0.567
Cs-137	0.084	0.007	0.012	0.009	0.054	0.016
Mn-54	0.558	0.068	0.017	0.047	0.060	0.165

Figure 1. Transfer factors of ^{60}Co, ^{137}Cs and ^{54}Mn from the investigated soils to orchard grass, after conditioning at two temperature regimes for one month

The TF values, obtained after one-month conditioning of the contaminated soil samples at temperate or hot temperature are shown in Figure 1.

The determined TF values are in agreement with the data for variation range of TF from soil to grass, published by the IAEA [7]. According to the literature, the mean value and minimum and maximum TF values are as follows:

^{60}Co (mean 7.7×10^{-2}, min 4.0×10^{-2}, max 1.7×10^{-1}) and ^{137}Cs (mean 6.3×10^{-2}, min 4.8×10^{-3}, max 9.9×10^{-1}) ^{54}Mn (mean 6.4×10^{-1}, min 1.1×10^{-1}, max 2.7), summarized for grass for all soil types [7].

The TF values for ^{137}Cs and ^{54}Mn from the present experiment are closer to the minimum value of the determined variation range, which shows low risk of their transfer within the food chain. The TF value for ^{60}Co in Gleyic fluvisol after conditioning at hot temperature, however, is found to be higher than the maximum value, reported by the IAEA. This might be due to the short period of aging, preceding the vegetation experiment, which led to weaker fixation of some radionuclides to the soil phases. The results, presented in Figure 1, show that the storage of the soils at high temperature caused decrease of the transfer of

^{137}Cs to orchard grass. This might be explained by the drying and wetting of the soil subsamples during conditioning, which have been found to increase the aging of Cs in soils from the Chernobyl and Mediterranean areas [11]. The authors have pointed out that the drying-wetting cycles accelerated the dehydration of the clay interlayers and subsequent interlayer collapse, which caused the Cs trapping and was responsible for the additional decrease in Cs desorption [11].

Decrease of TF values was found for the three radionuclides in Albic cambisol soil. This can be ascribed to the highest CEC and highest content of quartz and muscovite (Table 1), which favoured immobilization of the radionuclides. The studies of Manceau et al. [12] showed that Co co-precipitates on the quartz surface or precipitate as a neoformed trioctahedral of Co-rich clay. The high content of muscovite favors the immobilization of Cs, due to its ability to sorb strongly onto micaceous clay minerals [11]-[13]. Slight increase of TF values for ^{54}Mn and ^{60}Co were registered in Calcaric chernozem soil, as a result of hot temperature conditioning. Highest risk of increased bioaccumulation of ^{54}Mn and ^{60}Co after one month storage at 40 °C was detected in Gleyic fluvisol soil, which has lowest CEC and lowest contents of quartz and micaceous minerals.

3.2. Prognostic risk assessment for radionuclide transfer within the food chain

The obtained TF values were used to calculate the prognostic maximum specific activities of the studied radionuclides in the examined soil types, above which the meat and milk are harmful to be consumed, if obtained from livestock fed by grass from contaminated soils.

The values for maximum permissible specific activities of the radionuclides, used for the calculations, as described in 2.3., were 500 Bq/kg for meat and 200 Bq/kg for milk for each radionuclide, as recommended by [10]. The highest available values of F_m and F_f, presuming highest risk, were used from [7]. The dry weight of grass, taken daily by given animal species was evaluated to 12 kg for a caw, 1.5 kg for a goat and 1.2 kg for a sheep.

The obtained data [Bq/g] are presented on Figs. 2-4 for the studied soil types, conditioned at temperate and hot temperature regimes.

The present data exceed the maximum permissible limits of specific activities of radionuclides in the soils, as they do not take into account the additional radiation exposure of the population, received due to external exposure or ingestion through inhalation.

The data shown on Figure 2 indicate that the prognostic maximum permissible specific activities of the studied radionuclides in Albic cambisol soil are higher if the grass growth was preceded by one-month conditioning of the soil at 40 °C, as compared to the conditioning at 15 °C. The estimated maximum permissible specific activity in this soil is 2.5 times higher for ^{60}Co, 11.5 higher for ^{137}Cs and 8.2 times higher for ^{54}Mn after rapid warming, compared to the storage at temperate temperature regime.

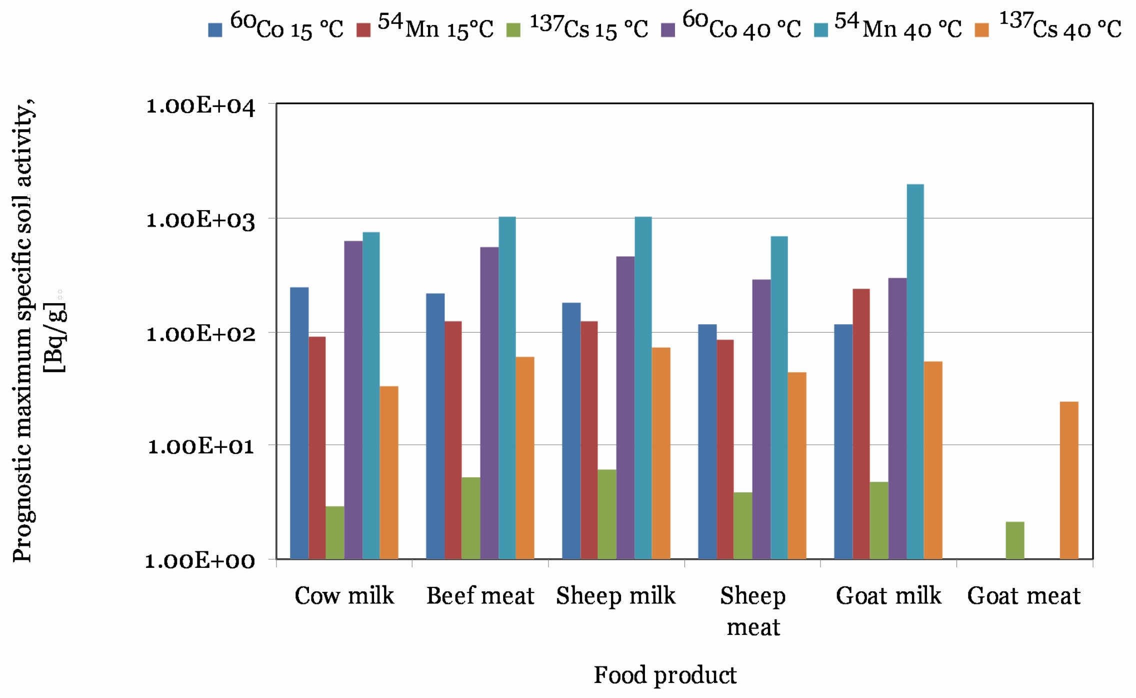


Figure 2. Prognostic specific activities of ^{137}Cs, ^{60}Co and ^{54}Mn in **Albic cambisol soil** [Bq/g], conditioned at two temperature regimes, above which the food products are harmful to be consumed

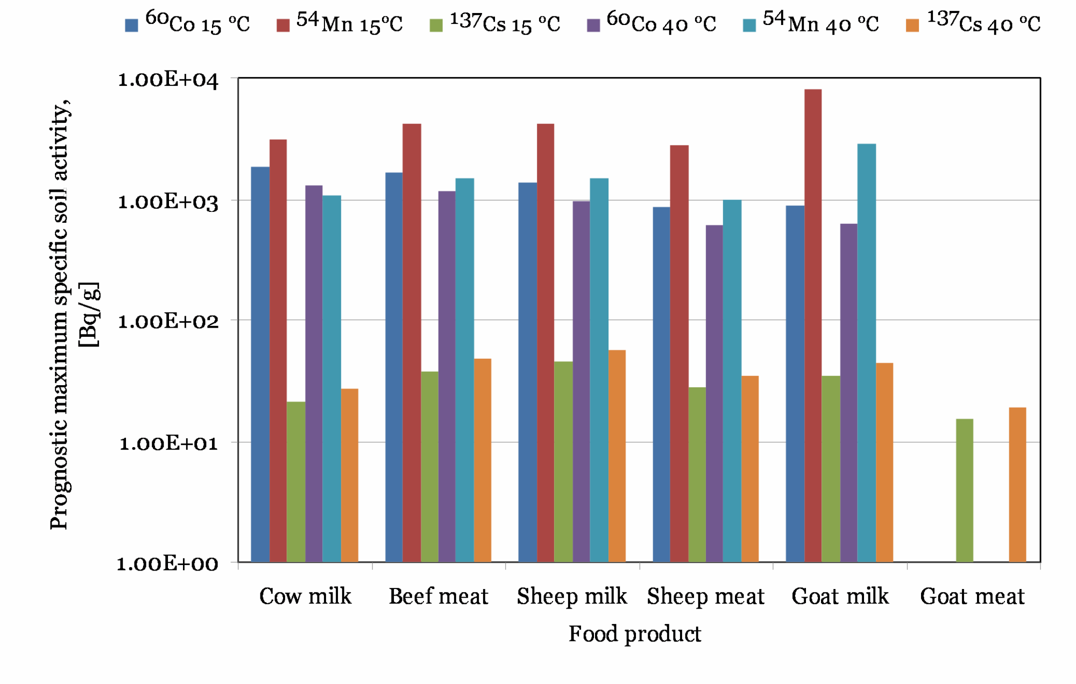


Figure 3. Prognostic specific activities of ^{137}Cs, ^{60}Co and ^{54}Mn in **Calcaric chernozem soil** [Bq/g], conditioned at two temperature regimes, above which the food products are harmful to be consumed

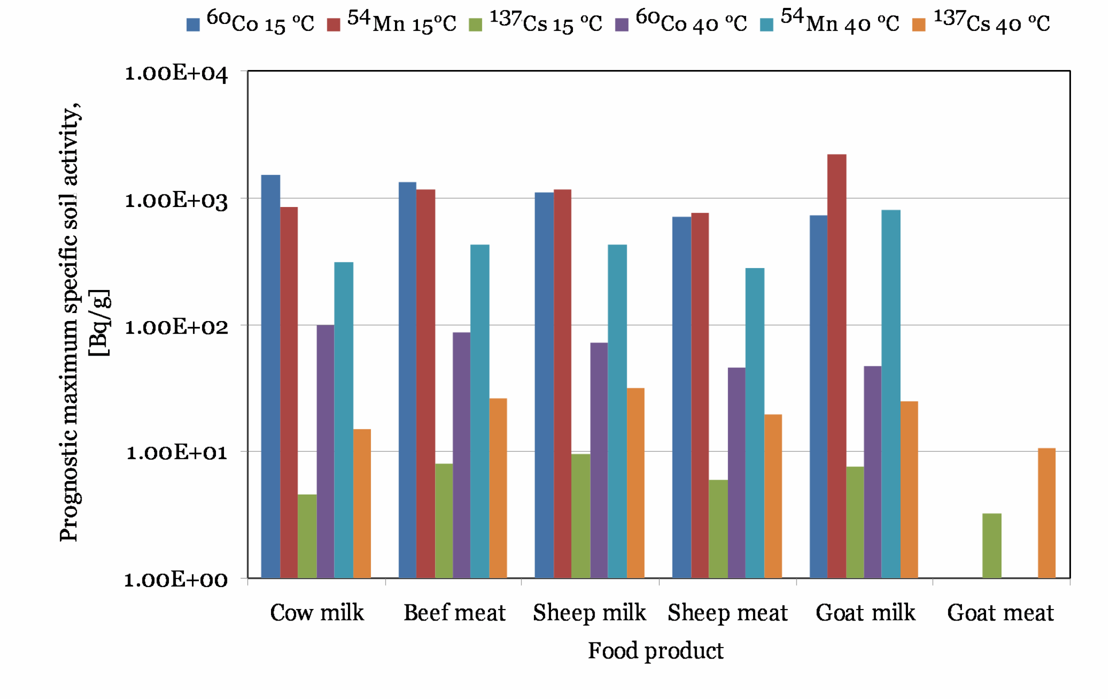


Figure 4. Prognostic specific activities of ^{137}Cs, ^{60}Co and ^{54}Mn in **Gleyic fluvisol soil** [Bq/g], conditioned at two temperature regimes, above which the food products are harmful to be consumed

The rapid warming for one month of Calcaric chernozem soil (Figure 3) reduces the prognostic maximum permissible specific activity of ^{60}Co (1.4 times) and ^{54}Mn (2.8 times) and slightly increases the maximum permissible specific activity of ^{137}Cs in this soil, above which the radionuclides transfer in the food chain exceeds the maximum permissible activities in meat and milk.

The conditioning of Gleyic fluvisol soil at hot temperature regime leads to the decrease of the prognostic maximum specific activity of ^{60}Co (15 times) and ^{54}Mn (2.8 times) and increases 3.3 times the maximum permissible specific activity of ^{137}Cs in this soil, estimated for harmless radionuclide transfer within the food chain (Figure 4).

The obtained data demonstrate the importance of the knowledge on the impact of temperature change on the transfer factors of radionuclides from different soil types to vegetation, when quick risk assessment of radioactive contamination is to be done.

4. CONCLUSION

The results from this study showed that rapid warming during one month after radioactive contamination influences the transfer of ^{137}Cs, ^{60}Co and ^{54}Mn from the soil to orchard grass. The temperature increase before the growing season was found to cause:

- Decrease of TF for radiocaesium, which was expressed more strongly in the Albic cambisol soil, which has highest content of micaceous minerals;
- Decrease of TF values of ^{60}Co and ^{54}Mn in the soil with high cation-exchange capacity, and highest content of quartz and muscovite;
- Increase of the TF values of ^{60}Co and ^{54}Mn to grass from the soil with low cation-exchange capacity and lower content of quartz and micaceous minerals.

Applicability of the data on the influence of sharp temperature change on the TF values for prognostic risk assessment after radioactive contamination was demonstrated by calculation of the maximum specific activity of the certain soil type, conditioned at given temperature regime, above which food products are harmful to be consumed.

Further studies on the influence of rapid changes of basic climatic parameters on the radionuclide transfer from different soil types within the food chain would contribute for more adequate risk assessment after radioactive contamination.

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