P-11-247

Transfer of long lived radionuclides in Chernobyl soils to edible plants

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Introduction

Transfer of long lived radionuclides such as ^{137}Cs, ^{90}Sr and transuranic elements in contaminated soils at Chernobyl to edible plants were examined. Analysis of radionuclides uptake from contaminated soils by plants is important from the point of view of not only the re-use of the soil, but also the decontamination of the contaminated soils by plants. This is especially true in the environment around the Chernobyl Nuclear Power Plants. The plant uptake experiments were made during 1996 and 1997 at Pripyat hothouse facilities belonged to Department of Radiology and Recultivation ChesCIR, Ukraine. The tests were carried out on sandy-podsolic soils and peaty soils which were adjusted in fixed levels of contamination by blending the same soil type of a different contamination density sampled in Chernobyl districts. In 1996, test cultivation was done in which two edible plants; Chinese cabbage and carrot were investigated on sandy-podsolic and peaty soils under two levels of radioactive soil contaminations. In 1997, four species of edible plant (radish and spinach are added) on sandy-podsolic and peaty soils under conditions of three levels of radioactive soil contaminations. The transfer factor is obtained by the ratio of concentration in plants (Bq/g-wet) to soils (Bq/g-dry) in this case. The IAEA summarized radionuclides uptake by plants (1,2). After the Chernobyl accident, many reports have been published concerning the radionuclides uptake from the contaminated soils by plants for ^{137}Cs and ^{90}Sr (for example; references 3). But, few reports have been published on plant uptake for the transuranic elements. This situation is not only in Chernobyl cases, but also in general (4,5).

Here, we examined the radionuclides uptake by Japanese plants from contaminated soils, not only for ^{137}Cs and ^{90}Sr,but also for transuranic elements. During 1996 and 1997 in CheSCIR in the Department of Radioecology and Recultivation, the joint Ukrainian - Japanese experiment was conducted aiming the study of the peculiarities of radionuclides transfer to the agricultural plants. In particular, the level of influence of soil and species factors upon the accumulation of the radionuclides by the agricultural plants (*Lactuca sativa L., Spinacia oleracea L., Raphanus sativus L., Daucus carota L*) was studied. Also the parameters of radionuclides transfer to the econimical parts of the plants (to the root-crops of radish (*Raphanus sativus L.*), carrot *(Daucus carota L.*) and leaf-crops of Chinese cabbage (*Lactuca sativa L.)*, spinach (*Spinacia oleracea L.*) on the soils of different density of radioactive contamination and of different typological belonging to were investigated. Here, we report the outline of the experiment.

Methods

In order to achieve the indicated above objectives and tasks, under the circumstances of closed soils (the green-house) the experimental artificial allotments were made (the concrete chutes filled with the soil of type needed and definite density of radioactive contamination). Two types of soils were used in the experiment (podsolic and peat); each type was divided into three levels of radioactivity (according to ^{137}Cs):

- First degree - up to 10 Ci/km^2 (0.37 TBq/ km^2; 2 Bq/g-dry soil)(the lower levels)
- Second degree - up to 100 Ci/km^2 (3.7 TBq/ km^2; 30 Bq/g-dry soil) (the middle levels)
- Third degree - up to 200 Ci/km^2(7.4 TBq/ km^2; 60 Bq/g-dry soil) (the higher levels)

Each variant of the experiment was laid in double repetition. The square of the variant was equal to 8 m^2. The square of crops for the separate type on the variant was 1.92 m^2.
The soils under experiment were investigated in many-sided manner from the agrochemical point of view. For measuring of pH of salt extraction from soil the potentiometrical method was used with soil : solution (1N KCl) 1:2.5 ratio. Hydrolytic acidity was measured by 0.1N NaOH filtration equal volume of extract solution, which was obtained by treatment of sample by 1N CH_3COONa, ratio soil : solution equals 1:2.5. Humus content was measured by the method of oxidation of soil carbon with mixed solution 0.4N $K_2Cr_2O_7+H_2SO_4$. Assessment of Ca and Mg was carried out with atomic absorption spectrometry involving direct aspiration of the aqueous solution 1N KCl into air-acetylene flame. Assessment of based-hydrolyzed N was carried out by interaction of aqueous solution 1N NaOH and soil by Kornfield method. Assessment of mobile phosphorous was measured by photocalorimeter of soil extraction (aqueous solution 0.2N HCl, soil: solution ratio - 1:5. Assessment of mobile K was measured by photocalorimeter of soil extraction (aqueous solution 0.2N HCl, soil: solution ratio - 1:5. Trace and toxic metals were assessed by atomic absorption spectrometry (soil extraction 0.1N HNO_3 solution, soil:solution ratio - 1:5.

Preparation for the sowing and the sowing itself

The preparation for the sowing included the putting the mineral fertilizers by the norm of $N_{60}P_{60}K_{90}$ (Table 1) according to the active substance, crumbly of soil and watering.

Table 1. The type and quantity of the fertilizers applied during the experiment

Kind of fertilizer	Amount of fertilizers, g/m^2
NH_4NO_3	17.4
$Ca(H_2PO_4)_2H_2O$	30.8
K_2O-30%	30.0

The distance in the lines between the crops was determined proceeding from the length of the line and the quantity of the sowing material. The sowing of the seeds of all the crops was conducted on the depth of 1 to 3 cm. The temperature of the air in the hot-house at the time of sowing was in the range of 32 - 34 C°.

Agrotechnical arrangements

The main purpose of the soil cultivation when growing up the vegetable crops under the conditions of the closed soils was the creation of the favorable conditions for the growth and development of the crops. The cultivation of the soil, in part the before-spring loosening as well as the post-spring one regulates the biological and chemical processes that are taking place under the conditions of closed soils.

Carrot:
The care of the crop includes the loosening of the soil before the sowing and during the vegetation period (the first loosening took place in the 3 - 4 leaves phase). The watering took place every 5 - 7 days. The first loosing of the distance between the lines took place for the depth of 4 - 6 cm when the plants had the height of 5 - 6 cm, the following ones were done for the depth of 10 cm.

Spinach and Chinese cabbage:
On the 4^{th} or 5^{th} day after the sowing the seeds were watered putting 250 - 300 m^3/ha or 5 - 6 l of water for the variant (the square of the variant - 1,62 m^2, then the watering took place every 1 - 2 days using the same quantity of water.

Radish:
The loosing of the distance between the lines took place every 10 - 12 days. The prevention of the extra concentrating the soil aim of such arrangement has been the aim of such arrangement, because this factor causes the plant to give the «arrow» and is the reason of forming the ugly and wooden roots.

The growth and the development of the plants

The development of all the plants, excluding spinach, was satisfactory though a little bit lifeless, which could be explained by the range of the reasons. It is known that spinach is the frost-hardy plant and can stand the frosts of 6 - 8 C° below zero, so the most favorable temperature for the spinach growth is about 15 - 18 C°, and under the conditions of the closed soil (under our conditions) the average day air temperature has been about 33 - 34 C°. Under the heighten temperature the spinach creates the small-sized leaves and quickly makes arrows, the same results are obtained when growing this plant under the conditions of the prolonged day. The given factors influenced the development of carrot especially during the first phase of ontogenesis.

Applying the standard methods of the registration of the growth and development of the plants during the systematic phenological observations the sproutness of the vegetable crops was determined at all the experimental variants. In our point of view the representative indicators that characterize the development of the plants at every stage of ontogenesis are the height of the plants, the size of the leaves and their coloring.

Pretreatment of samples and determination of radioactivities

The samples which were previously divided to the overhead and subterranean parts, were weighted and scrupulously washed by the distilled water, after that they were grated and dried under the temperature of 65 C° during 48 hours. Then they were grated to the powder and measured for their weights and radioactivities of the ^{137}Cs content in the plant samples on γ-ray spectrometer which is constantly intercalibrated.

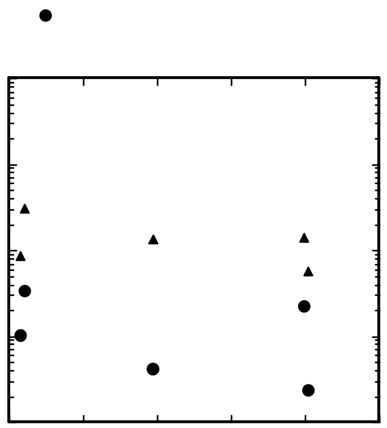
The determination of ^{90}Sr is accomplished by monitoring the Cherencov radiation from ^{90}Y in a liquid scintillation counter. ^{90}Y is the decay product of ^{90}Sr. Chemical yield of ^{90}Y is determined by adding a known amount of stable yttrium carrier. Interfering nuclides such as uranium, thorium, radium and their decay products as well as isotopes of caesium, potassium and strontium are separated from the sample by di-2-ethylhecsyl phosphoric acid (HDEHP). The main steps in this method are: Drying and ashing, Leaching, Solvent extraction of ^{90}Y by HDEHP, and Cherencov counting of ^{90}Y.

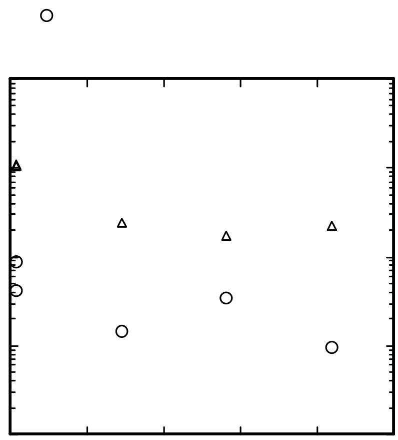
1. Drying and ashing: Soil samples were dried up to air-dried state, then were sieved through sieve with cell of 1 mm diameter and mixed thoroughly. Then in this samples (1-5g) was added carrier of stable yttrium and samples were ashed at temperature 610°C for 15 hours. The plant samples were dried at 95-105°C for 24, and then are milled and mixed. Samples were ashed up to white colour, at temperature 310°C for 3 hours and at temperature 610°C for 15 hours.
2. Leaching: Soil samples (1-5g) and plant ashes (1-2g) with added carrier of stable yttrium were leached with 1M HCl several times. All solutions were collected together.
3. Solvent extraction of ^{90}Y by HDEHP: ^{90}Y was extracted from water solution by HDEHP at pH1.0-1.2 and re-extracted by 3M HNO_3.
4. Cherencov counting of ^{90}Y: Cherencov counting of ^{90}Y was carried out with Quantulus alfa- beta-spectrometer.

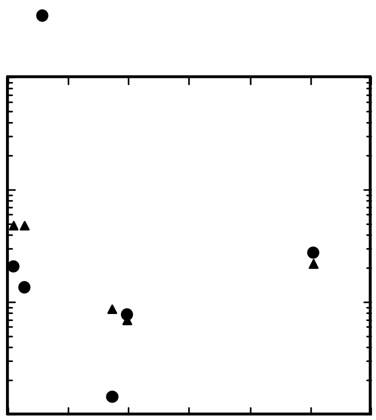
The determination of Pu is accomplished by monitoring the α–ray spectrometry. The dried sample is ashed under the temperature of 450°C during 24 hours. Chemical yield of Pu is determined by adding a known amount of ^{242}Pu. Ashed sample is leached with 8M HNO_3 (HF adding for soil sample). The leaching solution is added with Fe carrier and NH_4OH to co-precipitate the TRU. The precipitation of $Fe(OH)_3$ is solved with HCl and the solution is separated for Pu by an ion-exchange method. Pu source of α-ray spectrometry is prepared by an electro-deposition.

Results

Figure 1 and 2 show the transfer factors of ^{137}Cs and ^{90}Sr along with the soil concentrations, respectively. The transfer factor, which is the ratio of concentration in plants (Bq/g-wet) to soils (Bq/g-dry) in this case, ranged 0.03-1.1 for ^{137}Cs, 0.07-7.7 for ^{90}Sr in Chinese cabbage leaves, and 0.002-0.09 for ^{137}Cs, 0.01-0.7 for ^{90}Sr in the carrot roots, respectively. Obtained transfer factors are summarized in Table 2. These values are within the previously reported ones (1,2,4,5). As a whole, transfer factors of ^{137}Cs and ^{90}Sr in edible plants are larger in sandy-podsolic soils than in peaty soils. Dependence of transfer factor of ^{137}Cs on soil concentrations which has been reported in the previous report (6) was not so clear under the examined conditions in the vicinity of the Chernobyl districts as shown in these Figures, but has same tendency for ^{137}Cs with the reported one (6). This is partly because of the different range of the examined soil concentrations (This report : 2-60Bq/g-soil; Ref 6: 1-10 Bq/g-soil). Another reason may be attributed to the characteristics of radioactivity in the soils used. The radioactivity in the examined soils here in the present report were originated from hot particles, because they were sampled in the Ukrainian vicinity of Chernobyl NPP.







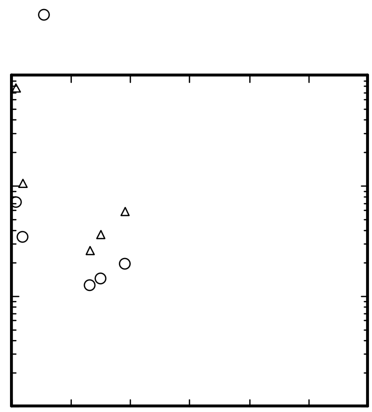


Table 2 Transfer factor for ^{137}Cs and ^{90}Sr (g-dry/g-wet)

	^{137}Cs	^{90}Sr
Chinese cabbage	0.03-1.1	0.07-8.
Spinach	0.009-0.1	0.06-9.
Carrot	0.002-0.09	0.02-0.7
Radish	0.008-0.04	0.05-2.

Table 3 Transfer factor for $^{239+240}Pu$ (g-dry/g-wet)

Soil	Radioactivity level (Bq/g dry)	carrot	Chinese cabbage
Peaty	Low (0.005)	0.04	0.02
	High (1.0)	0.0006	0.0006
Sandy	Low (0.009)	0.009	0.02
	High (0.8)	0.003	0.0005

Table 3 shows the transfer factors of Pu. Transfer factors ranged 0.0005-0.02 for $^{239+240}Pu$ in Chinese cabbage leaves, and 0.0006-0.04 for $^{239+240}Pu$ in carrot roots. As a whole, transfer factors of $^{239+240}Pu$ in edible plants are comparable in sandy-podsolic and peaty soils. Dependence of transfer factor of $^{239+240}Pu$ on soil concentrations of the radionuclide was found under the examined conditions, as shown in the Table 3.

Obtained transfer factors are larger than the previously reported values (1,2,4,5). This may be attributed to the chemical forms of Pu in the examined soils. Pu in the soils in the Chernobyl exclusion zone is hot particle oriented which are fine particles of nuclear fuel (7). The hot particles in the soils have been aged and eloded gradually (7,8). Then, some parts of Pu in the soils exist in water soluble, exchangeable, and organic forms which is associated with humic substances(9,10). These fractions of Pu are accessable to plant uptake. This may be one reason of higher transfer factors of Pu in the exclusion zone.

Summary

Transfer of long lived radionuclides such as ^{137}Cs, ^{90}Sr and $^{239+240}Pu$ in contaminated soils at Chernobyl districts to edible plants were examined under a controlled condition. The experiments were made during 1996 and 1997 at Pripyat hothouse facilities belonged to Department of Radiology and Recultivation ChesCIR, Ukraine. The tests were carried out on sand-podsolic soils and peaty soils which were adjusted in fixed levels of contamination by blending of the same soil type of a different contamination density sampled in Chernobyl districts. In 1996, test cultivation was done which two edible plants were investigated; Chinese cabbage and carrot on sand-podsolic and peat soils under conditions of two levels of radioactive soil contaminations. In 1997, four species of edible plant (radish and spinach are added) on sandy-podsolic and peaty soils under conditions of three levels of radioactive soil contaminations were examined, respectively.

The transfer factor, which is a ratio of concentration in plants (Bq/g-wet) to soils (Bq/g-dry) in this case, ranged 0.03-1.1 for ^{137}Cs, 0.07-7.7 for ^{90}Sr, 0.0005-0.02 for $^{239+240}Pu$ in Chinese cabbage leaves and 0.002-0.09 for ^{137}Cs, 0.01-0.7 for ^{90}Sr, 0.0006-0.04 for $^{239+240}Pu$ in carrot roots, respectively. These values are within the previously reported ones. As a whole, transfer factors of ^{137}Cs and ^{90}Sr in edible plants are larger in sand-podsolic soils than in peat soils. Transfer factors of ^{137}Cs and ^{90}Sr in edible plants are larger in sand-podsolic soils than in peat soils. Dependence of transfer factor of ^{137}Cs on soil concentrations of radionuclides which has been reported was not so clear in the examined ranges in the vicinity of the Chernobyl districts, but for Pu such tendency was found. As a whole, transfer factors ranged 0.0005-0.02 for $^{239+240}Pu$ in Chinese cabbage leaves, and 0.0006-0.04 for $^{239+240}Pu$ in carrot roots. These obtained transfer factors for $^{239+240}Pu$ are larger than the previously reported values. This may be attributed to the chemical forms of Pu in the soils examined.

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