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Executive Summary

Long-term field experiments have been carried out in the Chernobyl Exclusion zone to determine parameters describing technetium (^{99}Tc) transfer into five food plants (Lettuce, Radish, Wheat, Bean, Potato) from four types of soil, namely Podzoluvisol, Greyzem, Phaozem and Chernozem. Technetium, as ^{99}Tc, was added to the soils under field conditions in pertechnetate form. There was little effect of soil type in the first two years on Tc uptake by the plants. In the first and second year after contamination, Concentration Ratios (CR defined as ^{99}Tc activity concentration on crop (dry weight) divided by that in soil (dry weight)) of ^{99}Tc for radish roots and lettuce leaves ranged from 100 – 150 for the recently administered contamination whereas that for potato tubers (CR range 0.4-2.3) was two orders of magnitude lower than for radish and lettuce, and that for summer wheat grain was even lower at 0.8±0.3. After 6-7 years, root uptake of ^{99}Tc in wheat decreased 3-7 fold (CR was 0.02±0.01 to 0.14±0.03) and between 11-18 % of the total ^{99}Tc added remained in the upper 20 cm layers of the soils. The time taken for half of the added ^{99}Tc to be lost from the 20-cm arable soil layer due to its vertical migration and transfer to plants was rather low at c. 2-3 years. Climate conditions may have influenced technetium uptake by the plants as increases in the daily temperature were associated with increases in ^{99}Tc uptake by crops.

The long-term field experiments have been carried out in the Chernobyl Exclusion zone in order to determine the parameters of transuranic elements (TUE) (^{241}Am, ^{238,239,240}Pu), cesium (^{137}Cs) and strontium (^{90}Sr) transfer into plants from four types of soil, namely, Podzoluvisol, Greyzem, Phaozem and Chernozem. In frame of the task data on the experiment were compiled and a draft of article was prepared.

Under the same conditions, the highest CR values of the transuranic elements in radish roots, lettuce leaves, beans pods and wheat straw were observed in Podzoluvisol. For other soils the concentration ratios in each crop were similar, taking into account uncertainties of measurement.

CR of TUE in radish roots and lettuce leaves were close and can be roughly estimated by a value of $n \cdot 10^{-2}$ for Podzoluvisol and by a value of $n \cdot 10^{-3}$ for other soils. In general, the concentration ratios of plutonium and americium in beans pods varied in the range of 0.002-0.005 for Podzoluvisol and 0.0003-0.0018 for other soils. The lowest values of CR were obtained in wheat grains and potatoes tubers. A difference between americium and plutonium accumulation by wheat grains was observed. Accumulation of plutonium by wheat grains was a little higher in most cases than accumulation of americium. In general, transfer of americium and plutonium to grains for all soils can be estimated by CR values of $(2.4 \pm 1.5) \cdot 10^{-5}$ and $(7.0 \pm 5.0) \cdot 10^{-5}$, respectively. Concentration ratios of these radionuclides in potatoes for all soils varied in the range from 0.00006 to 0.0006 for plutonium and in the range from 0.00003 to 0.0004 for americium. Plutonium uptake by potatoes from Chernozem was in about an order of magnitude greater than that for americium.

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Uptake of technetium-99 by food crops under field conditions

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Introduction

The experiments that carried out in frame of this task, were a continuation of earlier works in the Chernobyl exclusion zone to reveal changes of transfer with time due to the radionuclide possible "ageing" to improve the dynamic modeling of soil-to-plant transfer for the long-lived radionuclides Tc-99, Pu, Am,. The work will include planting wheat at the experimental CEZ site, measurements of radionuclide activity and determination of the model parameters.

Uptake of technetium-99 by food crops under field conditions

Technetium is considered to be highly mobile in oxidized environments (Wildung et al., 1986). Due to its long half-life and high environmental mobility ^{99}Tc is a potential contributor to future radiation doses to humans and other living organisms, and is a particularly important radionuclide with respect to radioactive waste management. Therefore, there is considerable current interest in ^{99}Tc behaviour in the environment, including the study of its mobility in soil and its bioavailability that determines its uptake by plants and animals. Such information is essential for informed assessment of the environmental impacts of terrestrial nuclear storage facilities.

The extremely high mobility of Tc as a pertechnetate chemical species is an important issue for radioprotection purpose due to its high rate of transfer in the food chain. The CR observed values needs to be better discriminated according to environmental factors as they play a major role in Tc distribution. Aging parameter and differences between field and controlled laboratory conditions issues also needs to be addressed. Our study aimed to quantify the distribution of ^{99}Tc in the profile of four different soil types and the uptake of ^{99}TcO^{4-} by different food crops categories (leafy veg., root veg., tuber, cereal, leguminous) over several consecutive growing periods in field conditions, and over long time periods (2005-2014). The use of field conditions and the duration of the experiment make it possible to take into account the interaction of environmental variables and to acquire CR values representative of real agricultural conditions.

Methods

The experiments were carried out at the experimental site, described in earlier papers on iodine and chlorine (Kashparov et al., 2005a, 2005b), which is located in the Chornobyl Exclusion zone. Four different, previously uncontaminated, soil types (imported from other areas in Ukraine) were used in a total of eight soil lysimeters (two per soil type) to study the root uptake of ^{99}Tc by different agricultural crops. Each lysimeter area was 2×2 m2 and the depth was 0.5 m. According to FAO (IUSS Working Group WRB, 2006), the four soil types are classified as Podzoluvisol, Greyzem, Phaozem and Chernozem. They were taken from the 20-cm arable layer of agricultural lands in various regions of Ukraine. Such soil types are characterized by a large variation in agrochemical properties which would be expected to lead to considerable variation in ^{99}Tc root uptake with soil type.

The radionuclide for these experiments was obtained from V.G. Kholopin Radium Institute (Russia) in a form of solid potassium pertechnetate ($K^{99}TcO_4$). This salt was dissolved in 1M nitric acid to obtain 216 ml of a stock radionuclide solution (Tc oxidation state +7) with 10 kBq ml^{-1}. To obtain a ^{99}Tc solution an aliquot of 14 ml of the stock solution was diluted with tap water to a volume of 10 litres. Eight equal portions of 10 litres of the ^{99}Tc solution were prepared. The first administration of ^{99}Tc occurred on 16 May 2005. The upper 20-cm layers of soil in each lysimeter were carefully mixed and 10 litres of the ^{99}Tc solution were sprayed onto the 4 m^2 lysimeter surface using a special garden-watering tool. The ^{99}Tc activity concentration in the 20-cm soil layer was nominally about 120 Bq·kg^{-1} dw (0.2 μg·kg^{-1}); but varied within the range of 100-130 Bq·kg^{-1} dw due to the different soil densities in the lysimeters (Table 3). The soils were then fertilized with inorganic fertilizer (NPK 16:16:16) added in quantities of 60 g m^{-2} into Podzoluvisol and Chernozem and 50 g m^{-2} into Greyzem and Phaozem. Then, careful triple mixing was carried out to the same depth to achieve a uniform spatial distribution of ^{99}Tc and fertilizer within the arable layer in each lysimeter. The final density of contamination with ^{99}Tc within each lysimeter was 35 kBq·m^{-2}.

The crops were then planted on the same day as the ^{99}Tc was administered according to the following scheme for each of the eight lysimeters:

Radish **(***Raphanus sativus* L. convar. *radicula*.**,** French Breakfast variety) seeds were sown over 2 m^2 at a depth of 1-2 cm in 4 rows (inter-row distance ≈ 20 cm);

Four potato tubers (*Solanum tuberosum* L.) were planted between the radish rows;

Lettuce (*Lactuca sativa* L.) seeds were sown in lysimeters over 2 m^2 at a depth of 1-2 cm in 4 rows (inter-row distance ≈ 20 cm);

Beans (*Phaseolus vulgaris* L.) seeds were planted in 2 m^2 at a depth of 4-5 cm in 2 rows (inter-row distance ≈ 50 cm, distance between seeds in the row ≈ 10 cm).

Wheat (*Triticum aestivum* L.**)** was sown in 2 m^2 at a depth of 5-6 cm according to agronomic recommendations for the amount of seeds.

About one year later ^{99}Tc (30.6 kBq·m^{-2}) was again added onto the lysimeter soils on 27 April 2006 in a similar manner as that described above. The reason for two consecutive applications of ^{99}Tc was that at the beginning of the experiment we were not sure that measureable activity concentrations of ^{99}Tc would remain in the root layer in the second year. Crops were planted after fertilization and homogenization of the soils in the same quantities as during the previous growing season, but with crop rotation. Beans were planted on 22 May when the temperature increased to that recommended for this crop.

The third stage of the experiment started on 18 April 2007. The contaminated soil within the lysimeters was dug up, fertilized and only two of the crops were planted (summer wheat and potato). The areas of planting therefore increased to 4 m^2 for wheat and 2 m^2 for potato.

After three years of the experiment the lysimeters were left fallow for a period of about 5 years without further treatment before wheat was planted again. During the fallow period different natural graminaceous plant species typical for each soil type colonized the experimental lysimeters. The final stage of the experiment started on 18 April 2012 and finished. After digging which incorporated the currently growing vegetation into the soil, followed by homogenising and fertilization (at rates used previously), the summer wheat was sown as before (over 4 m^2).

Sampling procedures

Crops and soils were sampled at the normal time of harvesting of the mature agricultural species. Vegetables and wheat were harvested when matured.

From each lysimeter contaminated with ^{99}Tc only the edible part of harvested vegetables was taken for analysis. Each lysimeter yielded one sample per crop type giving four replicates. Wheat stalks were cut down and sheaved. The sheaves were then threshed in the laboratory by hand and samples of straw and grain were collected for treatment and measurement. Soil samples were collected in the lysimeters under each species using a cylindrical sampler (diameter of 3.7cm). Soil samples in each lysimeter were collected by the “envelope” method (5 cores) down to a depth of 20 cm. In addition, after the first contamination procedure and at the end of the growing periods, different layers of soil were collected from the lysimeters contaminated with ^{99}Tc. The soil samples were taken with a tube-type sampler (Ø 5 cm) to the depth of 30 cm and then each core was divided into 10-cm segments. Soil layer samples were also collected before the second administration of ^{99}Tc.

Tc extraction methods

A simple method was used to extract ^{99}Tc from the soil samples developed in the laboratory of NUBIP of Ukraine. The samples were dried in an electrical oven at 105°C to a constant weight. Then they were carefully mixed and sieved through a 1 mm sieve, and 50 g aliquots were collected for further analysis. Pair of aliquots was collected from each sample. One of them was spiked with a known amount of ^{99}Tc and used for determination of chemical recovery. These sub-samples were charred in a muffle furnace at 450°C for 12 hours. After charring, each soil sub-sample was washed out into a 500 mL funnel using deionised water. Water was added to the suspension until the volume was 200 mL. The acidity of the suspension was then reduced to pH 2-3 with concentrated nitric acid. To leach ^{99}Tc from the sample the suspension was placed on a hotplate magnetic stirrer at 80°C for 1 hour and then was left overnight. Then liquid and solid phases of the suspension were separated by filtration through a paper filter. The solid residue was used for a second extraction and was then discarded. The filtrates obtained from the two extractions were combined and evaporated at 80°C to a volume of 50 mL. This solution was then passed down through a cation exchange column, packed with cationic resin KU 2-8 ("Azot" OSTCHEM, Ukraine). This step removed the cationic radionuclides (^{40}K, ^{137}Cs, ^{90}Sr) also present in the solution. The eluent was evaporated to 20 mL volume and concentrated ammonium was added to maximally increase the pH of solution. Then 4 mL aliquots were collected into scintillation vials and prepared for liquid scintillation counting by the addition of 10 mL of OptiPhase HiSafe (PerkinElmer, USA) cocktail.

Initial treatment of the plant samples varied for different species. All plant samples were carefully washed, peeled (potato), chopped (radish, wheat straw) and dried in an electrical oven at 105°C to a constant weight. Subsequently, all samples were ground and two 10 g aliquots were collected for further radiochemical ^{99}Tc extraction procedures, which were the same as those given for soil.

Separation of ^{99}Tc from soil samples that had been collected since 2012 was carried out using TEVA discs (Eichrom Technology, Inc.). After drying as described above, the four studied soils were weighed (100 g) into appropriate quartz crucibles and ashed in a muffle furnace at 450°C for 12 hours to combust the organic matter. Two sub-samples were prepared from each soil. Before ashing one sample was spiked with 10 Bq of ^{99}Tc for the chemical recovery determination. After cooling the sample was transferred into a 1000 mL glass beaker and 500 mL of 1M HNO_3 was added. Then ^{99}Tc was leached from the sample on a hotplate magnetic stirrer at 80°C for 4 hours and left overnight. Residual solid matter from the suspension was removed by filtration through a paper filter. The solid residue was discharged. The extract was placed into a 2000 mL glass beaker and 20 mL of 30% H_2O_2 was added. The extract was diluted with water until a final sample volume of approximately 2 L was achieved and its acidity was reduced to pH=2 using ammonium hydroxide. The sample was heated to about 90°C for 1 hour to oxidize Tc(IV) to Tc (VII), to destroy residual organic matter and excess H_2O_2. After cooling to room temperature the sample solution was filtered through the TEVA disc by gravity. Tc-99 was measured by liquid scintillation counting by adding the disc directly to the scintillation cocktail (OptiPhase HiSafe).

Wheat straw and grain samples (2012-2014) were dried in an electrical oven at 105°C to a constant weight. Then, two sub-samples of each sample were collected for subsequent ^{99}Tc separation. We used 20 g and 100 g sub-samples for the straw and grain samples, respectively. After ashing, the plant sub-sample was placed into a 500 mL glass beaker using 200 mL of 1M HNO_3. All following extraction steps were the same as above for the soil samples except the final sample volume, which for the plant sub-samples was about 1 L.

Measurement of ^{99}Tc activity concentration

The ^{99}Tc activity concentration in soil and plant samples was measured using a liquid scintillation counting system with a MARK-IIIM liquid scintillation counter (Tracor Analytic Inc.,USA). OptiPhase HiSafe was used as the scintillation fluid. For our measurement scheme the minimum detectable activity was estimated to be 0.1 Bq. Each sample was analyzed with and without a ^{99}Tc spike to determine chemical recovery. Total chemical recoveries of Tc in the used extraction techniques varied in the range 65-97% and activities of the measured samples were corrected to 100%.

All reported specific activities and derivative values were calculated on a dry weight (dw) basis.

Statistical treatment of the data

Values of CR and activity concentrations of ^{99}Tc in soil are given as arithmetic means and standard deviation of the four replicas. One-way analysis of variance was performed on the natural logarithms of the data with a significance level $p<0.05$. The Newman-Keuls test was then used for comparison of means.

Results and Conclusions

Mobility of Tc in the soil profile

The decrease of ^{99}Tc activity concentrations in the upper 30-cm layer of soil one year after administration varied from 25% to 50% of its initial content. Vertical migration of ^{99}Tc with soil water and, to a varying but lesser extent plant uptake, played important roles in ^{99}Tc removal from the upper soil layer. However, after 6-7 years of soil contamination with ^{99}Tc, 11-18 % of the total ^{99}Tc added in the two stages remained in the upper 20 cm layers of the soils, corresponding to a period of half-removal of 2-3 years (rates of removal – 0.35-0.23 years-1). These observed values are an order of magnitude lower than the median value of 2.6 years-1 estimated by Baes (1979) for the 0-15 cm soil layer using a model of the radionuclide movement in soil with percolating water. The range of values in Baes (1979) study covers 3 orders of magnitude depending on soil parameters and Kd values. Baes' estimate is conservative because it does not take into account Tc retention in the root zone by plants. Similarly, the value estimated by Hoffman (1982) is several times higher than in our experiment, although it was derived in a short term field experiment for the 0-15 cm soil layer.

Water soluble fractions of ^{99}Tc ranged from 21 to 30 % in 2006 and from 4 to 9 % in 2014 (see Fig. 1). Over a long period of about 8 years, part of the ^{99}TcO^{4-} was converted to an immobile form resulting in a significant decrease in its availability for root uptake.

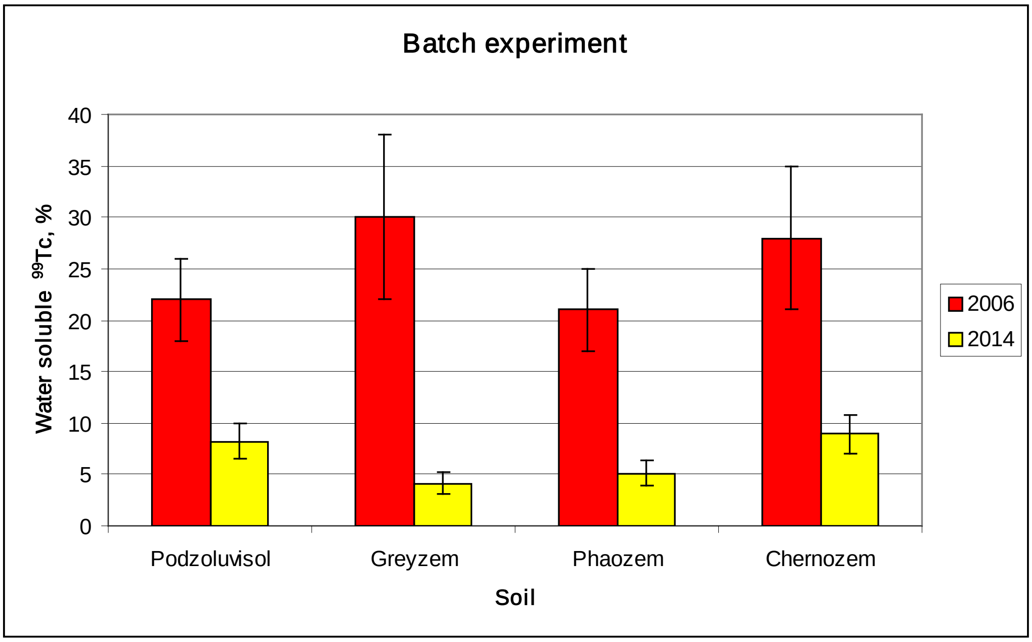

Fig.1 – Time dependence of water soluble fractions of ^{99}Tc during the experements

Plant uptake variation with soil type in field conditions

Tc accumulation by plants did not differ between soil types for the first two years after ^{99}Tc administration. A reason for the lack of difference could be the low rate of reduction of Tc(VII) to Tc(IV) under the experimental conditions. If this was the case, the difference that might be expected due to soil type may have been small compared with the possible influence of variation in environmental factors such as temperature and precipitation rate. Clearly, further experimental measurements would be needed to address these issues.

In subsequent years, when additional ^{99}Tc was not applied, accumulation of ^{99}Tc by the agricultural crops was higher from Podzoluvisol and Chernozem. Six years after the last administration of ^{99}Tc the biggest difference in CR values for summer wheat grain of about 6 fold, was observed between Podzoluvisol and Phaozem.

Effect of plant type on transfer

Under the experimental conditions, lettuce leaves and radish roots had the highest root uptake of ^{99}Tc added as pertechnetate. The CR for radish roots were in the range of 100-150 for freshly contaminated soils whilst those for lettuce leaves were in the range of 140-190. Uptake of ^{99}Tc in the potato tubers (CR range of 0.4-2.3) was two orders of magnitude lower than that in the two vegetables. Accumulation of ^{99}Tc by summer wheat grain was lowest and characterized by a CR range of 0.8±0.3 over the first 1-2 years.

Statistical analysis showed that uptake between plant types in 2005 was significantly different ($p<0.05$) from the same soil type except for lettuce and radish. In 2006, CR values for all crops growing on the same soil were also significantly different ($p<0.05$) except those for radish and green beans on Podzoluvisol, and lettuce and radish on all other soils. In 2007, the uptake between potato tubers and wheat grain growing on the same soil types were significantly different ($p<0.05$) except for the Podzoluvisol.

Time dependency

Hypothesis: ^{99}Tc uptake into plants will decrease rapidly with time in field conditions. This did not prove to be the case for summer wheat grain. Over a long period of about 6 years, part of the $^{99}TcO^{4-}$ was converted to an immobile form resulting in a significant decrease in its availability for root uptake. In 2012-2014, the CR values for wheat grain were one order of magnitude lower than those observed for fresh contamination (see Fig.2).

Climate conditions may significantly influence the plant uptake of technetium. Increases in the daily temperature may lead to enhanced uptake of ^{99}Tc by crops as previously observed by Echevarria et al. (2003).

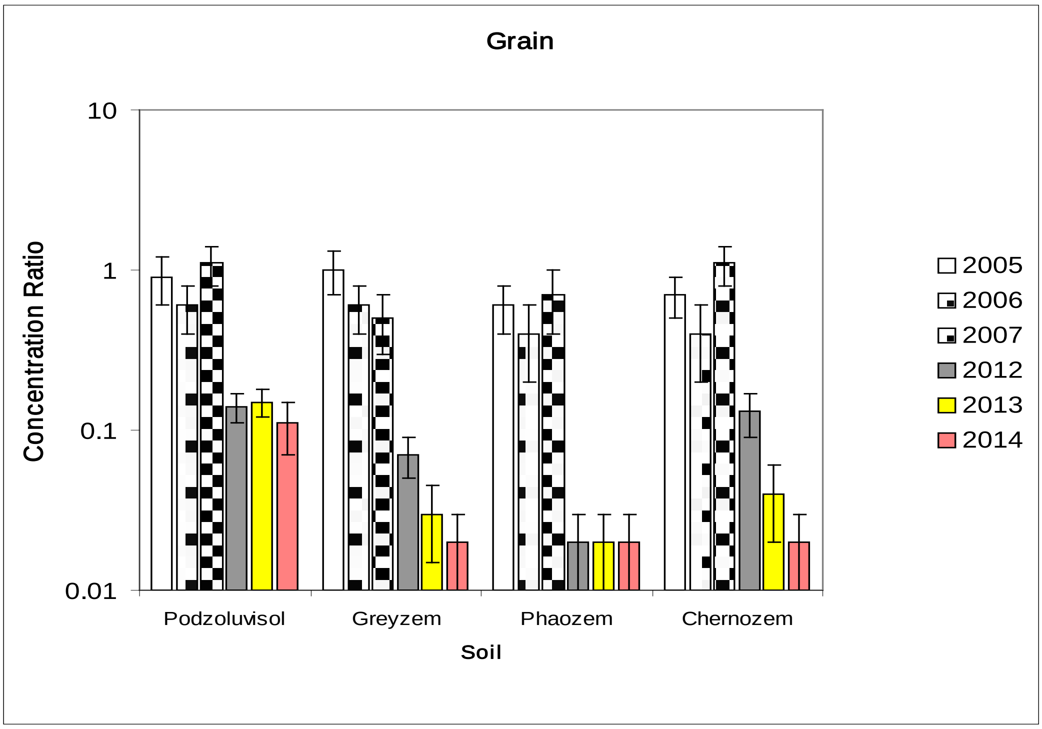

Fig.2 - Time dependences of ^{99}Tc concentration ratio for wheat grain

Plant uptake of transuranic radionuclides in field experiments

Methods

Site description

The experimental work was carried out at the site and for the same soil types, which had been used for previous study

The experimental site located at a distance of about 5 km from the ChNPP along the western trace of the Chernobyl radioactive fallout was selected because of its restricted territory status, it will be not used in the future because of the high terrestrial density of contamination with the long-lived radionuclides: ^{137}Cs -12±6 MBq/m^2; ^{90}Sr - 5±2 MBq/m^2; ^{238}Pu - 40±20 kBq/m^2; $^{239+240}$Pu - 90±30 kBq/m^2; ^{241}Am - 120±30 kBq/m^2. The soils studied were taken from the 20-cm arable layer of agricultural lands in the four regions of Ukraine, and then transported into the Chernobyl Exclusion zone and placed in four parcels, each parcel was 4x4 m in size and had a depth of 0.5 m. Each parcel was divided into four sub-parcels (2x2 m) by waterproof walls.

Radionuclides injection and fertilizing the soils

The transuranium radionuclides had been extracted by nitric acid from the high contaminated soil sampled in the Chernobyl zone. 1. On May 1, 2004**,** the radionuclides

^{241}Am and ^{238,239,240}Pu were injected in the soils. The solution of the radionuclides was sprayed onto the sub-parcel surfaces (4 m^2 each) using a special garden-watering tool. Thus, the following activities (on 01.05.2004) were injected in soil of each sub-parcel (2x2 m): ^{241}Am – 1.5±0.3 MBq, which corresponded to the terrestrial density of contamination of 0.38 MBq/m^2; ^{239,240}Pu - 0.32±0.06 MBq, which corresponded to the terrestrial density of contamination of 80 kBq/m^2 (about 1.5 and 0.3 kBq/kg for 20-cm soil layer, respectively).

Then, after some drying, the soils were mixed up to 20 cm depth by digging. The next step was fertilizing the soils and careful triple mixing to the same depth for homogenization of the radionuclides and fertilizers spatial distribution within the arable layer in each sub-parcel.

Also, a plot with the original local soil was prepared for cultivation of the crops within the experimental site. This soil was contaminated with Chornobyl radioactive fallout during the accident.

Planting the vegetables and treatments applied

Three successive crops of vegetables, summer wheat and potatoes were planted on the contaminated soils in the spring of each year from 2004 to 2006 and the crops grown to maturity before harvesting. Additional yield of summer wheat was obtained in 2007.

Sampling and analysis

Sampling of vegetables and soils was carried out at the time of harvesting of matured agricultural species. The sample volume depended on the vegetable yield from the sub-parcels. Usually only the parts of lettuce and potato yields were taken as the samples for measurement of radionuclides activities. Whole harvested amount of other vegetables were used as the samples because of their small yield. Soil samples were collected in the sub-parcels under each plant. Cylindrical sampler (diameter of 3.7 cm) was used for soil sampling. Soil samples in each parcel were collected by the “envelope” method (5 cores) to the 20 cm depth.

The radionuclides were isolated from pre-treated samples using standard radiochemical procedures (anion exchange chromatography) with following measurement by "Soloist" alpha-spectrometer with Soloist-U0300 detector ("EG&G ORTEC”).

Results and conclusion

Long term studies of the transfer of ^{238,239,240}Pu, ^{241}Am, ^{137}Cs and ^{90}Sr from different soils to the agricultural crops have been carried out using a field site in the Chornobyl Exclusion zone. Several crops have been grown at the site for tree years. Obtained experimental data on behavior of the radionuclides in soils and on their soil-to-plant transfer show that:

1. Under the same conditions (soil, period, plant) americium and plutonium uptake by radish roots, lettuce leaves, wheat straw and beans pods was quite similar. Though, CRs of these radionuclides in each crop were different. One can assume that radioactive contamination of these vegetative organs was not formed only by root uptake, and external contamination could play an important role. The plants could be contaminated by the

radionuclides during intensive rains when the rain splashes raise the small soil particles from the soil surface.

2. Accumulation of the radionuclides by the potatoes tubers and wheat grains was the lowest and caused by the root uptake. CR of plutonium in wheat grains for all soils slightly excided that for americium while concentration ratios in the peeled potato tubers were very close for both elements for all soils except Chernozem. Increasing of plutonium bioavailability in this soil can be caused by Pu-carbonate complexation.

3. Under the same conditions, the highest CR values of the transuranic elements in radish roots, lettuce leaves, beans pods and wheat straw were observed in Podzoluvisol. For other soils the concentration ratios in each crop were similar, taking into account uncertainties of measurement.

4. CR of the radionuclides in radish roots and lettuce leaves were close and can be roughly estimated by a value of $n \cdot 10^{-2}$ for Podzoluvisol and by a value of $n \cdot 10^{-3}$ for other soils. In general, the concentration ratios of plutonium and americium in beans pods varied in the range of 0.002-0.005 for Podzoluvisol and 0.0003-0.0018 for other soils.

5. The lowest values of CR were obtained in wheat grains and potatoes tubers. A difference between americium and plutonium accumulation by wheat grains was observed. In general, transfer of americium and plutonium to grains for all soils can be estimated by CR values of $(2.4 \pm 1.5) \cdot 10^{-5}$ and $(5.0 \pm 5.0) \cdot 10^{-4}$, respectively. Concentration ratios of these radionuclides in potatoes for all soils varied in the range from 0.00006 to 0.0006 for plutonium and in the range from 0.00003 to 0.0004 for americium.

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