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A comparative study of ^{40}K versus ^{137}Cs uptake as chemical analogs by vegetable plants at different concentrations of these nuclides in soil near the 30-km Chernobyl zone

V.I. Gaponenko*, N.V. Shamal and A.N. Nikitin

Institute of Radiobiology of the National Academy of Sciences of Belarus, 4 Fedyuninskogo St., 246007 Gomel, Belarus.

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Abstract – ^{137}Cs and ^{40}K uptake by vegetable plants was examined in a number of places in Belarus located at the outer perimeter of the 30-km Chernobyl zone. Differences in the uptake of ^{40}K and ^{137}Cs by plant species were not found to be related to the radionuclide content in the soil, but are essentially related to plant features. Vegetable plants were found to accumulate ^{40}K more actively than ^{137}Cs . For ^{40}K and ^{137}Cs , the ratio of the nuclide content in plants to their content in soil shows a tendency to decrease with increasing nuclide concentration in soil.

Keywords: ^{137}Cs / ^{40}K / vegetable plant / uptake / Chernobyl zone

1 Introduction

The Chernobyl nuclear power station accident in 1986 was known to result in contamination over large territories in Russia, Ukraine, and most seriously in Belarus with radionuclides (RNs). Nowadays, the ^{137}Cs -contaminated area is localized in the South-Eastern part of Belarus covering 1/5 of the country. A substantial part of the population in the Gomel region in Belarus lives in contaminated areas. Therefore, it is important to study the behavior of ^{137}Cs in the soil–plant system more precisely for radiation protection goals.

Potassium fertilizers are the main measure for decreasing radiocesium transfer into crops on contaminated soils, but fertilizers always contain ^{40}K . Interdependence of radiocesium and ^{40}K accumulation in crops after applying the fertilizers has not been studied in very great detail.

In studies of RN transfer into plants, the main attention is given to ^{137}Cs , ^{90}Sr and other artificial long-lived fission products of uranium and plutonium. A number of studies also include analysis of natural ^{40}K . Aleksakhin and Korneeva (1992) showed that radionuclide uptake is determined by a number of factors including the physicochemical properties of RNs, agrochemical characteristics of soil, biological features of plants and agricultural techniques. Moiseev *et al.* (1974) observed strong discrimination of ^{137}Cs transfer from soil to plants with respect to ^{40}K in experiments on wheat and oat. Discrimination coefficients for plant species and their parts were found to be different but independent of soil moisture within 30–60%.

A number of research groups evaluated the influence of various factors on ^{137}Cs uptake and accumulation by plants as

well as their relationship with the chemical analog, ^{40}K . Bunzl *et al.* (2000) studied the relationship between ^{40}K and ^{137}Cs in soil and plant samples from alpine pastures in Germany and Italy and found a significant negative correlation between the elements. They made the conclusion that radiocesium and potassium do not necessarily behave in an analogous way when a large variety of plant species is examined. Butkus and Konstantinova (2005) compared ^{137}Cs and ^{40}K uptake by plant species with various root systems (fern and grass) as well as by plants with no roots at all (moss) for soil in Lithuania. Fern was found to accumulate ^{137}Cs more effectively than grass and moss. ^{137}Cs and ^{40}K specific activity for fern was found to be higher than that for soil. Fern was also found to accumulate ^{137}Cs better than ^{40}K . Papastefanou *et al.* (1999) evaluated uptake parameters of fallout-derived ^{137}Cs along with naturally occurring RNs such as ^{40}K and ^7Be in soil and grass in northern Greece. Eleven years after the Chernobyl accident, the transfer coefficients from soil to grass were found to be 0.20 for ^{137}Cs , 0.73 for ^{40}K and 0.42 for ^7Be .

Simon *et al.* (2002) studied ^{137}Cs and ^{40}K uptake for 7 species of native plants on the Marshall Islands. The ^{137}Cs concentration in plants and soil samples varied in wide ranges (0.08–3900 Bq kg⁻¹), whereas the ^{40}K concentration in soil was within the range of 2.3–55 Bq kg⁻¹. No systematic variation of ^{40}K relative to ^{137}Cs was observed. For both ^{40}K and ^{137}Cs a decreasing concentration ratio (with increasing radioisotope concentration in soil) was observed.

Ehlken and Kirchner (2002) demonstrated that the radionuclide concentration in the soil is not the only factor influencing its uptake by a plant. A competition effect of major ions

* gaponen@imaph.bas-net.by

present in the soil-plant system plays a certain part in the rate of its accumulation.

As previously reported (Gaponenko *et al.*, 1988; Grodzinski, 1989; Gudkov, 1991; Aleksakhin and Korneeva, 1992; Parfenov and Yakushev, 1995; Gaponenko and Konoplya, 2007), plant species differences do occur in uptake and accumulation of RNs. These are determined by a number of factors including morpho-physiological characteristics of plants, and physicochemical and climate parameters of the environment. Dragović *et al.* (2004) examined ^{137}Cs activity levels in mosses from highland ecosystems of Serbia and Montenegro and found that biomolecules inside moss cells bind only 2.3–3.3% of the absorbed ^{137}Cs , whereas this value rose to 26.1–43.1% for cell membranes. A conclusion was made that this behavior has a chemical origin and cannot be interpreted as species-specific.

In our studies of radionuclide behavior in the soil-plant system after the Chernobyl accident in 1988, carried out in the Mozyr district, a negative dependence was found for the ^{137}Cs concentration ratio (CR) versus its activity in soil (Gaponenko and Konoplya, 2007). On a semi-logarithmic scale, the dependence obeys straight-line behavior. Notably, the dependence was found for a large number of plots for both cultivated and non-arable soil. Non-arable soil plots were examined in the exclusion zone of Chernobyl NPP.

The objective of this paper is to compare the uptake of artificial ^{137}Cs and natural ^{40}K radionuclides by vegetable crops from the soils contaminated due to the Chernobyl accident in the early and late periods of plant ontogenesis.

2 Materials and methods

2.1 Sample plot locations around the Chernobyl 30-km exclusion zone

The Gomel region is located in the South-Eastern part of Belarus bordering Russia and Ukraine; the total area is approx. 40 000 km², and the population is 1.6 million (1995). Since the Chernobyl accident in 1986, the population in the Bragin, Naroulya and Khoyniki districts has substantially decreased. Arable land covers 36% of the total area including plowed fields (20%), and grassland and pastures (13%). Cereal crops, leguminous plants and feed crops dominate in the cultivation area. In the Gomel region the crystalline basement is covered by sedimentary Proterozoic, palaios and anthropogenic layers. The area is flat ground.

All plots described in the present paper are located on the territory recognized to have radionuclide contamination from the Chernobyl accident fallout. According to Belarusian law, the polluted territory notion defines areas with ^{137}Cs contamination density of soil higher than 37 kBq m⁻². 57% of the overall ^{137}Cs -contaminated Belarusian agricultural land is located in the Gomel region. Nine places were examined in the Khoyniki (1–3), Bragin (4–7) and Naroulya (8–9) districts (Figure 1). All places are located at the outer perimeter of the Chernobyl 30-km exclusion zone. The radiological and chemical parameters of the soils are presented in Table 1.

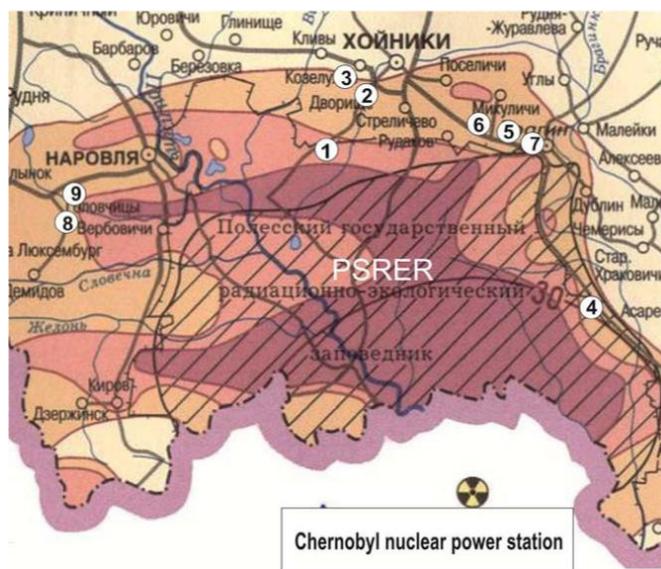


Figure 1. Map of sampling plots. 1- Novoselki, 2- Dvorische, 3- Sudkovo, 4- Savichi, 5- Burki, 6- Kovali, 7- Soboli, 8- Golovtchicy, 9- Buda Golovtchickaya.

2.2 Sampling of soil and plants

Soil and vegetable samples were collected at the sample plots and analyzed.

Soil samples were taken within the topsoil (20 cm) using a 248-cm³ sampler. For every plot, 5 soil samples were taken with subsequent drying, sifting (1-mm cell size) and homogenizing.

^{137}Cs and ^{40}K uptake was investigated in beet (*Beta vulgaris* L.), onion (*Allium cepa* L.), pepper (*Capsicum annum* L.), carrot (*Daucus sativus* (Hoffm.) Roehl.), potato (*Solanum tuberosum* L.) and vegetable marrow (*Cucurbita pepo* L., var. *giromontina*). For every plot coupled samples of soil and all plants were selected. Beet (leaves + root crops) and bulb onion (leaves) samples were taken in June, and pepper (fruits), beet (root crops), carrot (root crops), potato (tuber crops) and vegetable marrow samples were taken in September 2007. Plants were sampled from the selected plots in quantities enabling ^{137}Cs and ^{40}K determination 4 times. Samples were washed, crumbled and dried. The dried biomass was homogenized to enhance the reliability of the results.

2.3 Analysis of the chemical parameters of the samples

The soil type is sod-podzol. Assessing the chemical parameters of soil was carried out according to the procedures of the national standards (GOST) adopted in Belarus.

2.3.1 Soil pH (GOST 26483-85)

Exchangeable cations, nitrates and mobile sulfur were extracted from the soil samples by 1 M solution of potassium chloride. The ratio of soil and solution was 1:2.5. The pH of

Table 1. Characteristics of sampling plots.

Place	Specific activity of dry soil (Bq kg ⁻¹)		pH	Humus (%)	K ₂ O (exchange) ppm	P ₂ O ₅ (exchange) ppm	CaO (exchange) ppm
	¹³⁷ Cs	⁴⁰ K					
Novoselki	1831	196	6,34	3,9	458	1752	1293
Dvorische	1660	309	6,55	2,5	493	1146	1156
Sudkovo	664	376	5,21	2,9	663	1000	894
Savichi	539	223	6,43	3,2	422	1447	1433
Burki	1117	433	6,61	4,3	862	1728	1461
Kovali	1204	438	6,92	2,8	664	1582	1301
Soboli	1477	398	7,09	2,8	579	1403	1349
Golovtchicy	1272	114	6,40	3,5	473	821	892
Buda Golovtchickaya	758	63	6,12	2,7	252	489	762

the soil was determined by a potentiometer with a glass electrode. The total error of the method is 0.1 pH units.

2.3.2 Humus (GOST 26213-91)

The method is based on the oxidation of an organic substance by solution of potassium dichromate in sulfuric acid and subsequent trivalent chromium concentration measurement with a photoelectric colorimeter which is equivalent to the organic substance content. The maximal value of the relative error of the analysis is 10–20% ($p = 0.95$) depending on the percentage of organic matter in the soil.

2.3.3 Exchangeable calcium (GOST 26487-85)

Exchangeable calcium and magnesium were extracted from the soil with potassium chloride. Calcium was titrated by trilon B at pH 12.5–13.0; dark blue chromium acid was used as an indicator. Magnesium was titrated at a pH close to 10; dark blue chromium acid was used as an indicator. The total relative error of the method is 7.5–17%, depending on the amount of calcium in the soil.

2.3.4 Mobile compounds of phosphorus and potassium (GOST 26207-91)

Mobile phosphorus and potassium compounds were extracted from the soil with a solution of 2 M hydrochloric acid. The ratio of soil and solution was 1:5 for mineral horizons and 1:50 for organic horizons. Phosphorus content was measured with a photoelectric colorimeter in the form of blue phosphomolybdic complex. Potassium was determined by a flame photometer. The maximal value of the relative error of the analysis is 10–20% ($p = 0.95$) depending on the P₂O₅ percentage in the soil.

2.4 Analysis of the radioactivity of the samples

¹³⁷Cs and ⁴⁰K specific activity measurement was carried out using a Canberra Packard gamma spectrometer

with a coaxial Ge(Li) semiconductor detector with an advanced energy range. The measurement range of γ -radiation is 40–10 000 keV. The relative efficiency of spectrum registration at 1.33 MeV is 22.4%. The relative error of ¹³⁷Cs and ⁴⁰K specific activity measurements in samples ranged from 5 to 10% with respect to the sample activity. The lowest detectable activity for ¹³⁷Cs for acquisition time of 1 hour and 100 cm³ volume was 3 Bq.

For plant and soil radioactivity characterization, the specific activity (SA) value, in Bq kg⁻¹, was used. The dimensionless concentration ratio ($R = SA \text{ in plant} / SA \text{ in soil}$) was used as the measurement of ¹³⁷Cs or ⁴⁰K uptake by plants. The data on activity and concentration ratios for ⁴⁰K and ¹³⁷Cs CRs were determined as the mean values averaged over all plots.

3 Results and discussion

3.1 Chemical and radioactive parameters of the soils in sample plots

The soil in the sample plots is slightly acid or neutral, except for Sudkovo, where the soil pH is strongly acid. The humus content in the soil is very high in Dvorische, Sudkovo, Soboli, Kovali and Buda Golovtchickaya, and high in Novoselki, Savichi, Burki and Golovtchicy. The content of exchangeable potassium and phosphorus is high in all places except for Buda Golovtchickay, where it was very high. The content of exchangeable calcium is low in Buda Golovtchickaya, medium in Golovtchicy, Sudkovo and Dvorische, and high elsewhere.

Our measurements show that the density of soil contamination by ¹³⁷Cs (top 20-cm soil layer) in the sample plots is 140–500 kBq m⁻². The specific activity of ⁴⁰K in the top 20-cm soil layer varied from 15–63 Bq kg⁻¹ for one group of sample plots (Demidovo, Buda Golovchitskaya) to 398–438 Bq kg⁻¹ for another group (Soboli, Burki, Kovali). The specific activity of ¹³⁷Cs in the top 20-cm soil layer varied from 539–758 Bq kg⁻¹ (Savichi, Buda Golovchitskaya) to 1660–1831 Bq kg⁻¹ (Dvoryshche, Novoselki).

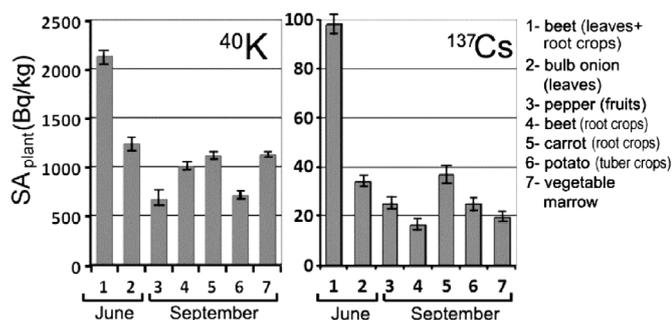


Figure 2. Specific activity of ^{40}K and ^{137}Cs in vegetable plants in private farmsteads in the Gomel area (2007, dry biomass).

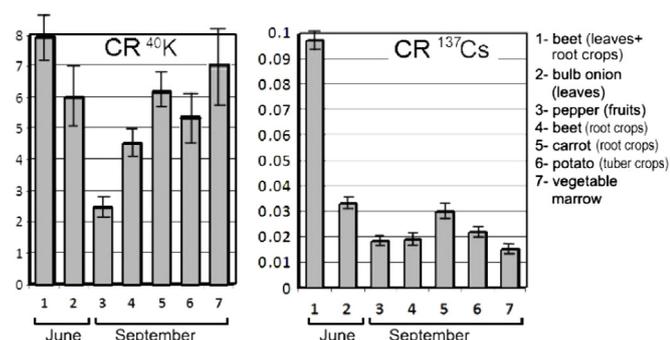


Figure 3. ^{40}K and ^{137}Cs concentration ratio for vegetable plants in private farmsteads in the Gomel area (2007, dry biomass).

3.2 Analysis of ^{137}Cs and ^{40}K uptake

Differences in RN accumulation among species were pronounced for vegetable plants examined with respect to the SA criterion both for the early and late periods of ontogenesis (Figure 2). At the beginning of June, the specific activity of beet plants with respect to ^{40}K was maximal (2133 Bq kg⁻¹), whereas the specific activity of bulb onion was 1.73 times lower. For these plants, the specific activity of ^{137}Cs was 98.0 and 34.0 Bq kg⁻¹ (natural moisture), respectively, *i.e.* lower than the permissible level (up to 100 Bq kg⁻¹ with natural moisture). For ^{40}K the permissible level is not regulated because of its natural origin.

In the late period of the vegetative season (the end of September), most of the vegetables had lower radioactivity in tissues. For example, for beet the specific activity of ^{40}K was 1003 Bq kg⁻¹, *i.e.* more than two times lower than in June. The SA in red pepper fruits, vegetable marrow, carrot root crop and potato tuber crop were within 671–1124 Bq kg⁻¹. In September, the specific activity of ^{137}Cs in beet plants was 6.16 times lower than in June. The SA of this radionuclide in the other vegetables ranged from 19.4 to 37.2 Bq kg⁻¹.

Notably, considerable differences also occur among vegetable plants with respect to the concentration ratio (Figures 3 and 4). This leads to the conclusion that differences in the uptake and accumulation of radionuclides by plant species are not related to the different concentrations of each nuclide in the soil.

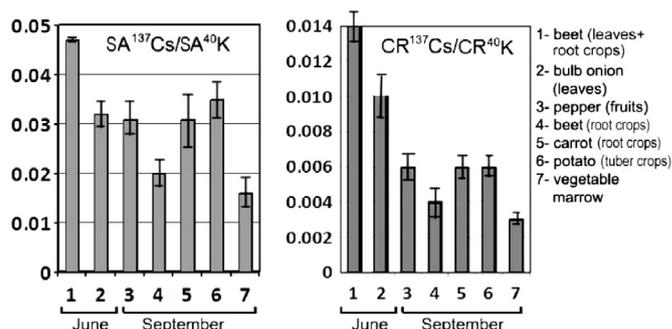


Figure 4. Ratio of specific activities of ^{40}K and ^{137}Cs in plants calculated based on data for dry biomass (left panel) and ratio of the CR of ^{137}Cs versus ^{40}K (right panel).

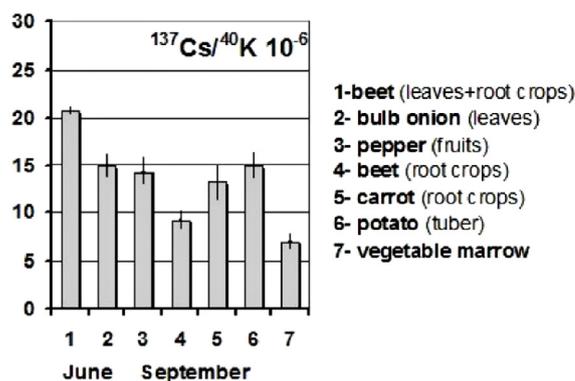


Figure 5. Ratio in moles of ^{40}K and ^{137}Cs contents in vegetable plants.

Furthermore, the values of the concentration ratio ($CR = SA_{\text{plant}} / SA_{\text{soil}}$) for ^{40}K are much higher than for ^{137}Cs . The ratio $CR^{40\text{K}} / CR^{137\text{Cs}}$ was lower in June as compared with September.

It is important to emphasize that for the full characterization of ^{40}K and ^{137}Cs as chemical analogs, it is useful to compare the ratios of these nuclides in plant tissues with respect to another criterion, namely in molar concentration. 1 Bq of ^{40}K is equal to 9.435×10^{-8} mole and 1 Bq of ^{137}Cs is equal to 4.258×10^{-11} mole.

We found (Figure 5) in June the highest content of both nuclides was found in beet plants: 201 $\mu\text{mol kg}^{-1}$ ^{40}K and 4.17 nmol kg⁻¹ ^{137}Cs , whereas the lowest values were found in bulb onion leaves, 115 $\mu\text{mol kg}^{-1}$ ^{40}K and 0.58 nmol kg⁻¹ ^{137}Cs . In September, these values were found to be considerably lower.

In summertime, the RN content evaluated in moles was higher than in fall, being equal for ^{40}K to 49.3–234 and 14.3–170 $\mu\text{mol kg}^{-1}$, respectively (dry biomass), and for ^{137}Cs : 0.58–5.23 and 0.17–2.02 nmol kg⁻¹, respectively. The molar ratio $^{137}\text{Cs}/^{40}\text{K}$ also had higher values in June as compared with September: $(4.28\text{--}28.5) \times 10^{-6}$ and $(2.1\text{--}17.8) \times 10^{-6}$, respectively. The radioactive isotopes ^{40}K and ^{137}Cs are only a small part of these elements in soil and biomass. However, we assume that the percentage of such isotopes in total storage in soil and plant biomass is the same. The comparison of

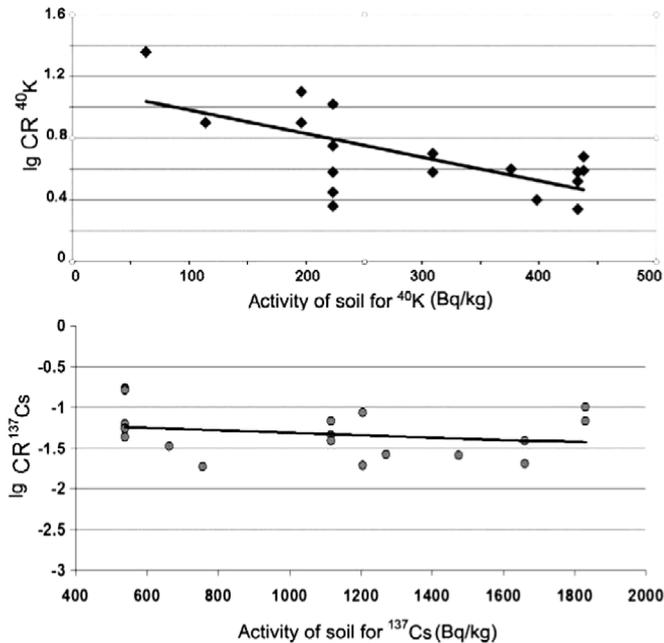


Figure 6. ⁴⁰K and ¹³⁷Cs uptake by vegetable plants versus their different concentrations in soil.

concentration ratios, expressed in moles, shows the most active isotope uptake by different plants more accurately.

Based on the data obtained, a conclusion can be made that ⁴⁰K ions enter vegetable plants more actively than ¹³⁷Cs. This can be explained by the stronger fixation of ¹³⁷Cs and the exceptional importance of potassium in biological systems. Though cesium ions are analogs of potassium ions, their ionic radii are 167 ppm and 133 ppm, correspondingly.

The CR of both nuclides in plants showed an inverse relationship with the concentration of Cs and K in soil (Figure 6). For ⁴⁰K, such a tendency is reported for the first time. The dependence features a semi-logarithmic character and is modeled by a linear fit, as has been described elsewhere (Gaponenko and Shamal, 2010).

4 Conclusions

Based on experimental studies for a number of plots at the outer perimeter of the 30-km Chernobyl zone, plant species differences in ⁴⁰K and ¹³⁷Cs uptake were established, and were not found to be related to the radionuclide content in soil, but are essentially related to plant features.

For ⁴⁰K and ¹³⁷Cs as chemical analogs, the activity, molar concentration and their ratio in vegetable tissues were evaluated. It was concluded that ⁴⁰K more actively enters plants than ¹³⁷Cs, which may result from weaker fixation of the former in soil and the exceptional importance of potassium for plant biochemistry and physiology.

The concentration ratio for ⁴⁰K and ¹³⁷Cs in plants to their content in soil shows a tendency to decrease with increasing nuclide concentration in soil. The tendency on a semi-logarithmic scale can be approximated by a straight line. For ⁴⁰K, such a tendency is reported for the first time.

Notably, the results obtained are not only of theoretical significance but also practically important, since they show that while the nuclide content in soil will decrease, its penetration into vegetables and other organisms including humans will rise. This should be taken into account when making a prognosis on the remediation of regions contaminated with radiocesium.

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