ASSESSMENT OF THE CONTAMINATION LEVEL OF A PODZOLIZED CHERNOZEM WITH NUCLIDES IN A LONG-TERM LAND USE

HRYGORII HOSPODARENKO, IHOR PROKOPCHUK*, OLGA NIKITINA,
VITALII LIUBYCH

Uman National University of Horticulture, Ukraine

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Prolonged systematic application of mineral fertilisers contributes to increasing the yielding capacity of agricultural crops. However, it can lead to significant changes in the composition, properties and formation of agricultural soil regimes. The findings of the research have shown that the application of mineral fertilisers leads to the change of radioactive nuclides content in the soil. The research was conducted under conditions of a long-term stationary field experiment (Uman, Ukraine), using different rates of mineral fertilisers $N_{45}P_{45}K_{45}$, $N_{90}P_{90}K_{90}$ and $N_{135}P_{135}K_{135}$. Soil samples (podzolized chernozem) were selected from the depths of 0–20, 20–40 and 40–60 cm. Specific activity of radionuclides was determined by the spectrometric analysis. Using experimental results we have demonstrated that under a long-term application (50 years) different rates of mineral fertilisers effect the specific activity of radioactive isotopes in the soil (^{226}Ra , ^{232}Th , ^{40}K , ^{137}Cs , ^{90}Sr). The specific activity of radionuclides in a podzolized chernozem and in winter wheat grain was established. Winter wheat plants accumulated ^{232}Th at the highest levels, but the use of fertilisers reduced it in a larger mass of the crop. The absorption of radioactive nuclides by winter wheat grain grown after peas and silage corn depending on fertilisation changed similar to growing it after clover as a previous crop. According to the data of specific activity of radioactive nuclides in the soil and winter wheat grain, the coefficient of their biological absorption was calculated.

Key words: radioactive isotopes, radium, thorium, potassium, cesium, strontium, specific activity of radioactive nuclides

At present, anthropogenic pressure on the environment is quite substantial. In particular, the use of fertilisers as part of the intensification of agricultural production gave rise to a number of new problems, making the ecological problem the most acute. Numerous studies have shown that different fertilisers have different effects on soil properties. They enter into complex interactions with the soil, leading to various transformations that depend on a number of factors: properties of fertiliser materials and soil, weather conditions, agricultural technologies, cultivated crops, etc. (Andrews *et al.* 2002;

Clark *et al.* 1998; Hřčková *et al.* 2018; Šimanský 2016). It is established that a long-term period of intensive agricultural use of the territories with high often unreasonable man-caused pressure has led to significant changes in the composition, properties and formation of arable soil regimes (Tobiašová *et al.* 2013).

However, the negative effect of fertilisers is often exaggerated. In the environmental sense, they cannot be equated with pesticides or man-made contaminants (Petrenko *et al.* 2017). The development of rational plant nutrition systems should include

Ihor Prokopchuk, Candidate of Agricultural Science, associate professor (*Corresponding author), Uman National University of Horticulture, 1 Institutska St., Uman, Cherkassy region, Ukraine. E-mail: pivotbi@ukr.net

the obligatory assessment of the possible negative impact on agrocenosis and the ways to minimize it. Knowing special features of sorption, migration and translocation of toxicants in the soil helps to avoid unwanted phenomena and provide high efficiency of fertilisers (Fateev & Zakharova 2005).

At present, the development of agriculture is impossible without the application of fertilisers which allow to increase soil fertility, yielding capacity and improve the quality of agricultural products. The yield increase is ensured by 50% due to their application (Hospodarenko 2018).

First of all, the negative influence of fertilisers on the environment is can be explained by their chemical composition and the presence of ballast substances (Begey & Shuvar 2007; Taylor *et al.* 2016). When applying fertilisers under each crop, it is necessary to take into account the maximum permissible concentrations of chemical elements in the soil.

Consequently, in agriculture, along with improving yielding capacity and product quality, the studies aimed at preserving and protecting the environment against technogenic pollution should be considered relevant. It is necessary to introduce environmental resource-saving technologies that would ensure the maintenance of clean soil, water and air (Begey & Shuvar 2007).

In the literature there is practically no data on the radioactivity levels of potassium fertilisers. It is known that 1g of potassium accounts for 29.6 Bq of ^{40}K . Radioactive potassium (^{40}K) has a half-life of 1.2×10^9 years. It is characterized by a beta-negative decay type with the energy of 1.32 MeV (88.4%) and partial gamma radiation (K-capture) with the energy of 1.46 MeV (11.6%) (Hussain & Hussain 2011). The content of ^{40}K in the soil can vary within a wide range (100–750 Bq/kg). Potassium isotopes are usually found in soil in a strongly bound form and have a low rate of transition to plants. In nature potassium exists in the form of three isotopes ^{39}K (93.1%), ^{41}K (6.9%) and radioactive ^{40}K (0.012%). Radiation of ^{40}K makes up only 12% in the general background of natural sources of radiation (Tzortzis *et al.* 2004).

The most dangerous radionuclides are ^{137}Cs and ^{90}Sr due to their active inclusion in the circulation of a trophic chain. The migration of these radionuclides in the soil-plant system leads to their accumulation

in plant products. To reduce the transition of ^{137}Cs into plant products, it is necessary to apply mineral (especially potassium) and organic fertilisers and to carry out the liming of soils. The analysis of the specific activity of ^{137}Cs and ^{90}Sr in the vegetative mass of buckwheat has shown that the level of their concentration is influenced by the soil acidity and fertiliser system. Thus, there is a decrease of the specific activity of ^{137}Cs in winter wheat grain by 18–30% in variants with organic-mineral fertiliser system. This is mainly due to its binding with organic substance (Trembitska 2010).

The aim of the research was to investigate the degree of nuclide pollution of a podzolized chernozem under a long-term land use in order to determine the transformation of radionuclides in winter wheat grain.

MATERIAL AND METHODS

The research was carried out on the experimental field of Uman National University of Horticulture in the stationary experiment of the Department of Agrochemistry and Soil Science. The experiment was launched in 1964, and it is based on a 10-field crop rotation extended in time and space (spring barley + meadow clover, meadow clover, winter wheat, sugar beet, corn, peas, winter wheat, silage corn, winter wheat, sugar beet). The object of the research was a podzolized clay-loam black soil of the experimental field. Mineral fertiliser system was used ($\text{N}_{45}\text{P}_{45}\text{K}_{45}$; $\text{N}_{90}\text{P}_{90}\text{K}_{90}$; $\text{N}_{135}\text{P}_{135}\text{K}_{135}$). Fertiliser rates were applied in the form of ammonium nitrate, granulated superphosphate, mixed potassium salt and potassium chloride. The total area of the plot was 180 m², the experimental plot covered 100 m², the experiment was repeated three times on the same location. Winter wheat variety Favoritka was cultivated. Wheat grain samples were taken at the stage of full maturity.

Before the experiment the soil was under a long-term cultivation under field crops. Soils samples taken before the experiment (1964) had the following parameters: content of physical clay – 66.5%, base saturation – 95%, humus content – 3.31%; content of easily hydrolysable organic nitrogen (according to the Tjurin – Kononova method); mobile com-

pounds of phosphorus and potassium (according to Chirikov method) – 122 and 135 mg/kg respectively; pH_{KCl} – 6.2. Soil was characterized by such indicators at the time of setting up the experiment.

The soil samples were taken by earth boring machine from the layers at the depth of 0–20, 20–40 and 40–60 cm during 2015–2017. The experiment had three repetitions. The coefficient of biological absorption (CBA) of heavy metals in grain was determined by the formula (Baron *et al.* 2006).

$$\text{CBA} = \frac{C}{E},$$

where: C – content of the chemical element in grain [mg/kg]; E – content of the element in the soil [mg/kg].

The method of spectrometric analysis was used to study specific activity of radionuclides (spectrometer – SEG-001, SEB-01-150). Statistical data processing was carried out by using the software Microsoft Excel 2010 and STATISTICA 10.

RESULTS

The application of fertilisers has rather active influence on the soil. The presence of various toxic impurities, unsatisfactory quality, as well as pos-

sible violations of the application technology can lead to significant negative consequences. Failure to adhere to scientifically grounded application measures of fertilisers, imperfect methods of their introduction may have a negative impact on individual components of the biosphere, on the state of the environment and on a man (Boukhenfouf & Boucenna 2011; Alshahri & Alqahtani 2015). The level of radionuclide accumulation is inversely proportional to the soil fertility level: the higher the fertility is, the lower is the soil resistance to radionuclide contamination (Howard 2000).

Data in Table 1 show that the specific activity of radioactive isotopes increases with an increase in the amount of applied fertilisers in the soil. The use of triple rate of fertilisers increased radionuclide content compared to the variant without the application of fertilisers: ^{226}Ra by 26%, ^{232}Th by 118%, ^{40}K by 30%, ^{137}Cs by 26% and ^{90}Sr by 6%.

As a result of the dispersion analysis, it was established that the impact of the previous plant on the content of radioactive nuclides was the highest since the partial eta-squared was 0.97 and the one of fertiliser was the smallest (0.46) (Figure 1). Important quantitative indicators are absorption completeness of radionuclides (sorption) and stability of their fixation in the absorbed state. For example, if we compare the stability of fixation of ^{90}Sr and ^{137}Cs in the absorbed state, it turns out that ^{90}Sr is displaced

Figure 1. The effect level of the investigated factors on the specific activity of radionuclides in the soil

in greater quantity than ^{137}Cs (so, absorbed cesium is firmly fixed). It is confirmed by the data obtained from the research. The stability of radionuclide fixation is uneven on different soils. They are fixed better in black soil.

In general, unlike strontium, cesium is better sorbed (fixed) by minerals, especially by clay. The radiation hazard of strontium is 6 times higher than that of cesium under the same level of soil contamination. In order to reduce the degree of migration, such agrochemical measures are carried out as liming acidic soils, poor in metabolic calcium and introducing organic fertilisers (humus, peat and manure). To reduce strontium uptake phosphorus fertilisers are used, and to reduce cesium uptake – potassium fertilisers.

The presence of two natural radioactive nuclides is characteristic for potassium fertilisers (^{226}Ra and ^{40}K). In this case, the main radionuclide is ^{40}K (Kondo *et al.* 2015). Radioactive potassium (^{40}K) is the

most common among the natural radioactive isotopes. Its general activity in the earth's crust is greater than the activity of all other isotopes taken together. It is widely distributed in soils, especially clay ones, where it is firmly retained as a result of sorption processes. Due to the content of ^{40}K (0.0119%) one gram of natural potassium is characterized by the activity of 29.6 Bq (Kaste *et al.* 2003).

The use of potassium fertilisers with residual natural radioactivity does not significantly increase the radioactivity of agricultural crops, but sometimes it increases by several times. Such phenomenon is usually observed in young plants under the application of high rates of potassium fertilisers (Becegato *et al.* 2008).

Specific activity of potassium and radium in the soil depended on the rate of potassium fertilisers. Specific activity of ^{40}K increased from 12 to 30% depending on the rate of applied fertilisers and the specific activity of ^{226}Ra increased from 11 to 26%.

T a b l e 1

Specific activity of radionuclides in the soil after long-term growing (50 years) of field crops using different fertilisers (2015–2017) [Bq/kg]

Experiment variant	Soil layer [cm]	Radionuclide				
		^{226}Ra	^{232}Th	^{40}K	^{137}Cs	^{90}Sr
Without fertilisers	0–20	25.8±0.5*	20.4±0.7*	105.1±4.1*	8.4±0.4*	3.1±0.05*
	20–40	20.1±0.6*	20.6±0.5*	77.3±2.8*	7.2±0.4*	2.0±0.04*
	40–60	14.9±0.8*	20.1±0.9*	52.3±1.2*	4.2±0.3*	1.8±0.02*
$\text{N}_{45}\text{P}_{45}\text{K}_{45}$	0–20	28.6±0.5*	30.4±0.8*	118.0±3.7*	9.0±0.2*	3.2±0.03*
	20–40	22.3±0.6*	20.6±0.5*	81.2±1.9*	7.4±0.4*	2.0±0.04*
	40–60	15.2±0.7*	19.6±0.7*	56.4±4.2*	4.5±0.4*	1.8±0.03*
$\text{N}_{90}\text{P}_{90}\text{K}_{90}$	0–20	30.7±0.8*	38.6±0.6*	128.1±4.4*	9.9±0.3*	3.1±0.03*
	20–40	24.8±0.9*	26.3±0.5*	95.9±3.6*	7.6±0.3*	2.2±0.04*
	40–60	15.2±0.9*	20.1±0.6*	58.7±1.6*	4.6±0.3*	1.9±0.05*
$\text{N}_{135}\text{P}_{135}\text{K}_{135}$	0–20	32.6±0.7*	44.6±0.5*	136.2±1.7*	10.6±0.3*	3.3±0.04*
	20–40	27.1±0.7*	30.6±0.3*	110.0±3.0*	7.8±0.3*	2.4±0.03*
	40–60	15.1±0.3*	20.8±0.7*	61.2±3.2*	4.5±0.4*	1.9±0.05*

Note: * – insignificant variation (V = 0–10 %)

Downward the soil profile their specific activity decreased and at the depth of 4,060 cm it was 2 times lower than in the upper layer of 0–20 cm.

Safety of potassium fertilisers was established in a long-term field experiment. Potassium radiation, creating natural radiation background, is not limited by current norms and is not dangerous to human health.

Application of fertilisers in the field crop rotation changes the specific activity of radioisotopes in winter wheat grain (Table 2).

When cultivating winter wheat after silage corn, specific activity is lower compared to other previous crops (clover and peas). The specific activity of ^{232}Th in winter wheat grain was lower compared to ^{40}K (18.1–22.8 Bq/kg) depending on the rate

of fertilisers and previous plants. Also, it has been found that ^{232}Th absorption was higher under growing wheat after clover and peas (19.6–22.8 Bq/kg of grain) than under growing wheat after silage corn (18.1–19.8 Bq/kg).

Specific activity of ^{226}Ra was almost unaffected by the investigated components of agricultural technologies of winter wheat cultivation. Its activity varied within a small range (6.0–6.8 Bq/kg).

^{90}Sr and ^{137}Cs radioisotopes are easily included in the migration processes in agricultural chains because their chemical properties are similar like those in calcium and potassium, that play an important role in the biosphere.

^{137}Cs and ^{90}Sr specific activity in winter wheat grain was the lowest compared to other radionu-

T a b l e 2

Specific activity of radionuclides in winter wheat grain using different fertilisers in the field crop rotation and preceding crops (2015–2017) [Bq/kg of dry mass]

Experiment variant (Factor A)		Radioactive nuclide				
		^{40}K	^{232}Th	^{226}Ra	^{137}Cs	^{90}Sr
Clover as a preceding crop (Factor B)						
Without fertilisers (control variant)		68.1 ± 1.5*	19.6 ± 0.5*	6.5 ± 0.03*	1.7 ± 0.06*	0.8 ± 0.04*
$\text{N}_{45}\text{P}_{45}\text{K}_{45}$		69.3 ± 2.3*	19.5 ± 0.2*	6.4 ± 0.04*	1.7 ± 0.05*	0.8 ± 0.04*
$\text{N}_{90}\text{P}_{90}\text{K}_{90}$		74.5 ± 1.3*	20.2 ± 0.4*	6.6 ± 0.03*	1.9 ± 0.05*	0.9 ± 0.03*
$\text{N}_{135}\text{P}_{135}\text{K}_{135}$		87.2 ± 1.5*	22.8 ± 0.5*	6.7 ± 0.03*	2.1 ± 0.03*	1.0 ± 0.04*
Peas as a preceding crop						
Without fertilisers (control variant)		68.4 ± 1.9*	19.5 ± 0.7*	6.6 ± 0.03*	1.7 ± 0.04*	0.8 ± 0.02*
$\text{N}_{45}\text{P}_{45}\text{K}_{45}$		70.1 ± 1.4*	19.8 ± 0.7*	6.7 ± 0.04*	1.8 ± 0.06*	0.8 ± 0.04*
$\text{N}_{90}\text{P}_{90}\text{K}_{90}$		76.2 ± 1.6*	20.4 ± 0.4*	6.7 ± 0.02*	1.9 ± 0.04*	1.0 ± 0.04*
$\text{N}_{135}\text{P}_{135}\text{K}_{135}$		86.4 ± 2.2*	21.7 ± 0.3*	6.8 ± 0.03*	2.1 ± 0.02*	1.1 ± 0.04*
Silage as a preceding crop						
Without fertilisers (control variant)		68.3 ± 1.4*	18.1 ± 0.7*	6.0 ± 0.03*	1.7 ± 0.05*	0.8 ± 0.02*
$\text{N}_{45}\text{P}_{45}\text{K}_{45}$		69.8 ± 1.8*	18.2 ± 0.7*	6.1 ± 0.03*	1.7 ± 0.06*	0.8 ± 0.03*
$\text{N}_{90}\text{P}_{90}\text{K}_{90}$		71.7 ± 1.4*	19.3 ± 0.5*	6.1 ± 0.03*	1.7 ± 0.06*	0.9 ± 0.03*
$\text{N}_{135}\text{P}_{135}\text{K}_{135}$		77.6 ± 1.4*	19.8 ± 0.4*	6.1 ± 0.03*	1.8 ± 0.03*	1.0 ± 0.02*
$LSD_{0.05}$	A	1.7	0.5	0.2	0.1	0.1
	B	1.4	0.4	0.2	0.1	0.1
MPC		–	–	–	80	140

MPC – maximum permissible concentration; $P = 0.002$

Note: * – insignificant variation ($V = 0–10\%$)

T a b l e 3

Coefficient of biological absorption of radioactive nuclides by winter wheat grain after a long-term application of fertilisers (50 years) in the field crop rotation and preceding crops

Experiment variant (Factor A)		Radioactive nuclide				
		⁴⁰ K	²³² Th	²²⁶ Ra	¹³⁷ Cs	⁹⁰ Sr
Clover as a preceding crop (Factor B)						
Without fertilisers (control variant)		0.65	0.96	0.25	0.20	0.26
N ₄₅ P ₄₅ K ₄₅		0.59	0.64	0.22	0.19	0.25
N ₉₀ P ₉₀ K ₉₀		0.60	0.55	0.22	0.20	0.28
N ₁₃₅ P ₁₃₅ K ₁₃₅		0.64	0.51	0.21	0.20	0.30
Peas as a preceding crop						
Without fertilisers (control variant)		0.65	0.96	0.26	0.20	0.27
N ₄₅ P ₄₅ K ₄₅		0.60	0.66	0.24	0.20	0.26
N ₉₀ P ₉₀ K ₉₀		0.61	0.57	0.23	0.19	0.31
N ₁₃₅ P ₁₃₅ K ₁₃₅		0.64	0.49	0.22	0.19	0.32
Silage corn as a preceding crop						
Without fertilisers (control variant)		0.66	0.95	0.25	0.21	0.29
N ₄₅ P ₄₅ K ₄₅		0.63	0.71	0.22	0.19	0.27
N ₉₀ P ₉₀ K ₉₀		0.60	0.61	0.22	0.18	0.28
N ₁₃₅ P ₁₃₅ K ₁₃₅		0.59	0.51	0.21	0.18	0.31
LSD _{0.05} by factors	A	0.02	0.03	0.01	0.01	0.02
	B	0.01	0.02	0.01	0.01	0.01

 $P = 0.002$

clides, correspondingly from 1.7 to 2.1 and from 0.8 to 1.1 Bq/kg, depending on the agricultural technology of cultivation. This indicates that a long-term application of fertilisers in the field crop rotation at mentioned rates is safe. At the same time, ¹³⁷Cs specific activity was 40–47 times and ⁹⁰Sr specific activity was 127–145 times lower than the maximum permissible concentration.

It is found that the application of potassium fertilisers contributes to a significant reduction of radioactive cesium content in the main as well as non-market part of wheat yields compared to the control variant (Pettigrew 2008). Potassium fertilisers are an effective and economically justifiable measure to reduce ¹³⁷Cs and ⁹⁰Sr penetration in plant products.

A long-term application of potassium fertilisers increases the radioactivity of the soil due to the

content of ⁴⁰K and ²²⁶Ra. However, the important ecological function of potassium is antagonism in relation to radioactive ¹³⁷Cs and ⁹⁰Sr. Many scientists note the significant role of potassium fertilisers in reducing the intensity and migration of radionuclides in the soil-plant system. Potassium inhibits ¹³⁷Cs transfer into crops of agrocenoses. The greatest effect is achieved with the application of high rates of potassium fertilisers (Aleksakhin *et al.* 1992; Hospodarenko 2018).

According to the data of specific activity of radioactive nuclides in the soil and in winter wheat grain presented in Table 3, the coefficient of biological absorption (CBA) of radionuclides was calculated (Table 3).

Winter wheat plants accumulated ²³²Th most of all. However, the use of fertilisers reduced it in a larger mass of the crop. Thus, in the variant

without fertilisers, the coefficient of biological absorption of ^{232}Th by winter wheat grain after clover was 0.96 and in the mineral fertiliser system it was 0.51–0.64 or less by 33–47%. Winter wheat grain accumulated the smallest amounts of ^{137}Cs , ^{90}Sr and ^{226}Ra .

The absorption of radioactive nuclides by winter wheat grain after peas and silage corn depending on fertilisers varied similar to that after the clover as a preceding plant.

The results of the research are ambiguous when compared to the similar studies that were published. Thus, in research (Fateev & Zakharova 2005), specific activity of ^{137}Cs under the application of $\text{N}_{90}\text{P}_{60}\text{K}_{90}$ decreased from 188.2 Bq/kg to 119.5 Bq/kg. In studies (Tsybul'ka *et al.* 2014) conducted on peat soils, the application of potassium fertilisers reduced the activity of this radioisotope in wheat grain by 20–30%, but the application of N_{60-120} increased it by 4–9%. It is obvious that specific activity of radionuclides in wheat grain varies not only after the use of fertilisers, but it also depends on the level of soil fertility created by long-term fertilisation.

CONCLUSIONS

Absorption of radionuclides by topsoil prevents them from moving through its profile and further penetration into groundwater. Thus, specific activity of radionuclides was two times lower at the depth of 40–60 cm than in the soil layer of 0–20 cm. This indicates that radioisotopes are fixed in the upper layers of the soil which in its turn increases their penetration into crop products.

The level of ^{137}Cs uptake by winter wheat plants from the soil is the highest, while the level of ^{90}Sr uptake is the lowest. The coefficient of biological absorption of ^{232}Th is significantly reduced with the improvement of the conditions of plant growth and plant development. The absorption of ^{40}K from the soil is significant but it is safe for human health. Contamination of crop products with radionuclides depends on the type and properties of soils, types, forms and rates of fertilisers and the duration of their application. Therefore, the radionuclide concentration in plants on different soils in different soil-climatic zones may be different at the same level of pollution.

Long-term application of potassium fertilisers increases the soil radioactivity due to the content of ^{40}K and ^{226}Ra but this radiation is safe for human health. In addition, potassium has an important ecological function being the antagonist in relation to radioactive ^{137}Cs and ^{90}Sr .

The analysis of the specific activity of radionuclides in winter wheat grain indicates that a long-term application of fertilisers (50 years) in the field crop rotation at the indicated fertiliser rates is safe.

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