THE CONTENT OF NATURAL RADIOACTIVE ISOTOPES IN SOIL OF KHARKOV REGION

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The content and migration of natural radioactive isotopes in some lot of the surface soil of the Kharkiv region have been studied by the help of low background gamma spectroscopy. The migration series of radioactive elements (Ra > U > Th) in the soil of the Kharkiv region is determined, which corresponds to the humid type of climate. A high radium mobility as respect to uranium and thorium has been found. A high correlation between ^{40}K vs ^{232}Th was found. Therefore, as a result of low mobility of thorium, it is possible to assume that thorium and potassium enter into the composition of the mother minerals. Correlation of ^{137}Cs with other radioactive isotopes was not significant in the soil of the Kharkiv region. This apparently indicates specifies in that a high mobility clay minerals can sorbing of cesium.

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1. INTRODUCTION

Soil radioactivity is formed by geochemical processes in the biosphere. The parent rock generally determines a concentration of radioactive elements in soil. In Tab. 1 shows typical concentrations of natural radionuclides in different soil types. Biological processes in the formation of soils lead to a greater accumulation of radionuclides in chernozems compared to gray forest or sod-podzolic soils. The leaching and precipitation of radionuclides by soil waters determine the uneven distribution of nuclide activity along the depth of the soil. For example, uranium has a high concentration in the deep layers due to the washing out of soluble fractions from the surface horizons, and thorium, on the contrary, tends to increase in the surface layers due to low solubility and greater weathering of other components from the soil surface. Potassium tends to a greater extent to surface layers as it characterizes by relatively high biological activity (see Table 1) [1].

Many radionuclides, especially analogs of calcium and actinides, have osteotropic properties and are being detected in the human body. This particularly applies to radium isotopes. Radium and its compounds are widely distributed in nature and are a major source of natural radiation. During the weathering, and migration of radium from rock to soil, it can 'move out' in the particulate phase, to be transported and deposited as loess, silt placers, and tertiary soil. However, that part of radium which tends to be solubilized in water – either with a ground or river water – moves along with the water stream until it is finally deposited in the soil through chemical or biological action. The range of ^{226}Ra concentrations for soil in normal areas varies from 3.7 to 125.8 Bq/kg [2]. It is known that the content and distribution of radium in soils affects the radon emanation.

Table 1. *The concentration of natural radionuclides in soils, Bq/kg*

Type of soil	^{238}U	^{232}Th	^{40}K
Serozems	31.5	48.1	666
Gray-brown	27.8	40.7	703
Chestnut	26.6	37	555
Chernozems	21.6	35.9	407
Gray forest	17.8	26.6	370
Sod-podzolic	15.2	22.2	300
Podzolic	8.9	12.2	148
Peaty	6.3	6.3	89

Thorium exists in soils mainly as thorite ($ThSiO_4$), monazite [$(Ce,La,Th,Nd)PO_4$], and thorianite (ThO_2). Although they are very resistant after extreme weathering, monazite and thorite end up being dissolved and then precipitates as thorianite.

Our planet displays the results of an unintentional, multibillion-year experiment between biota and Earth materials. One of the exciting frontiers in the geosciences is the investigation of how biota impacts chemical and physical processes in the Critical Zone. Understanding the feedbacks between life and Earth materials is important for society as well, because humans, a relatively recent addition to the

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Earth's biota, are increasingly testing the resilience of the Critical Zone. The response of Critical Zone processes to these stresses will ultimately impact the whole of the Earth's biosphere, and thereby, humans themselves [1].

The biotechnological introduction of microbial genes coding for toxic proteins against insects, weed species or other pests in the genome of crop plants is another important topic in the study of the environmental and health consequences of protein–mineral interactions in soil. Some of the best known and most controversial genetically modified plants are those containing the Bacillus thuringiensis (Bt) toxin. Commercially available Bt plants include maize, cotton, potato, tobacco and rice. Nevertheless, the balance between these clear advantages in the short- and medium-terms and the possible pernicious effects in the longer term needs to be studied. With the introduction of toxins, such as Bt and an increasing number of other potentially valuable insecticidal molecules, the risk of the development of resistance increases. This could have major negative consequences for agriculture, equivalent to bacterial resistance to antibiotics in medicine. Thus, studies on the long-term fate and side effects of Bt toxins in soil need to be pursued [3].

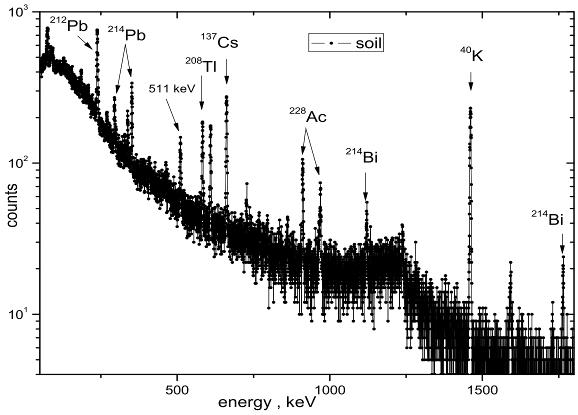

Fig.1. *The energy spectrum of soil sample*

Modern ideas about the formation of chernozems confirm the hypothesis of plant-terraneous origin. The most important processes of formation of chernozems are sodding and eluvial. The latter is expressed mainly in the profile migration of calcium bicarbonate, which is formed by the decomposition of plant remains rich in calcium. These processes develop under the perennial vegetation of grassy steppes in the forest-steppe and steppe zones under conditions of periodically washing and non-washable water regimes and form humus and carbonate profiles of chernozem [4]. Dust flux is greater than 10% of global sediment. Therefore the deposition from the atmosphere plays an important role in the biogeochemistry of soils worldwide [5].

The study of the content of ^{40}K, ^{232}Th, ^{238}U and their radioactive products in soils of a neighborhood of the Kharkiv region and an establishment of correlative connections between radioelements were the purposes of the present investigation.

2. MATERIALS AND METHODS

Samples of soils (15x15 cm^2 and h=10 cm) were collected in the neighborhood of Kharkiv on virgin soil or in the forest. Two samples were taken on arable land. Soil samples were dried at room temperature, then sieved through a 1-mm mesh to remove the roots of plants and stones and homogenized prior to analysis. Determination of the radionuclides in soil samples was performed by gamma spectrometer method on Ge(Li)-detector with the volume of 50 cm^3 and resolution of 3.2 keV at 1332 keV line. The measurement time of the gamma spectrum of each soil sample was about 24 hours. To reduce the influence of background, the detector is equipped with a three-layer Pb-Cu-Al protection [6] (Fig.1). The errors in determining the activity of radionuclides ^{40}K and ^{137}Cs do not exceed 20%. The same errors are found for ^{228}Ac, ^{212}Pb, ^{208}Tl from the family ^{232}Th and ^{214}Pb, ^{214}Bi (which is determined the parent ^{226}Ra) from the family ^{238}U. The more significant error is for ^{238}U, the activity of which was calculated from the activity of ^{235}U (line of 186 keV is equal sum's of lines from ^{226}Ra+^{235}U).

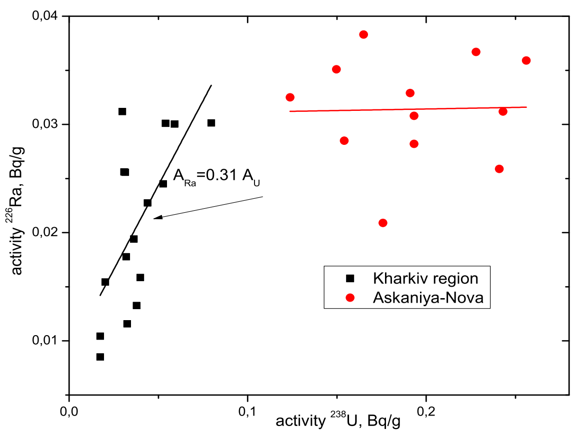

Fig.2. *Dependence of activity of ^{226}Ra vs ^{238}U in the soil of the Kharkiv region and Askaniya-Nova*

3. RESULTS AND DISCUSSION

It is known that the content of radioactive isotopes in the soil is largely determined by the parent rocks. However, environmental conditions have a significant effect on the migration of uranium, thorium and their decay products. For example, the migration of radium (an analog of calcium) depends most strongly on the environmental conditions. In Fig.2 shows the dependence of activity of ^{226}Ra on the activity of ^{238}U for various samples of soil from different regions of Ukraine. The data from Askaniya-Nova is taken from [7]. The Kharkiv region is characterized by a humid climate, which, apparently, determines the greater removal of radium from a given territory in the sea. Askaniya-Nova is characterized by a more arid climate. It can be assumed that the Askaniya-Nova region was often flooded with water (its height above sea level is 30 m). Therefore, the content of ^{226}Ra in the soil of Askaniya-Nova is somewhat higher than in the humid zone of the Kharkiv region. We note that in the soil, which is closer to the parent rock, there is a ^{226}Ra/^{238}U equilibrium. In our case, the content of radium for the Kharkiv region is much smaller (see Fig.2).

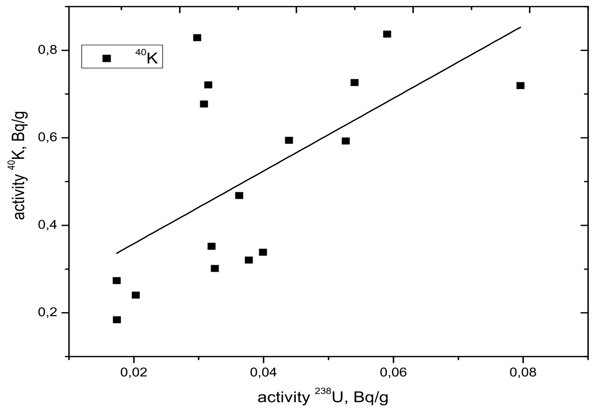

Fig.3. *Dependence of activity of ^{40}K vs ^{238}U in the soil of the Kharkiv region*

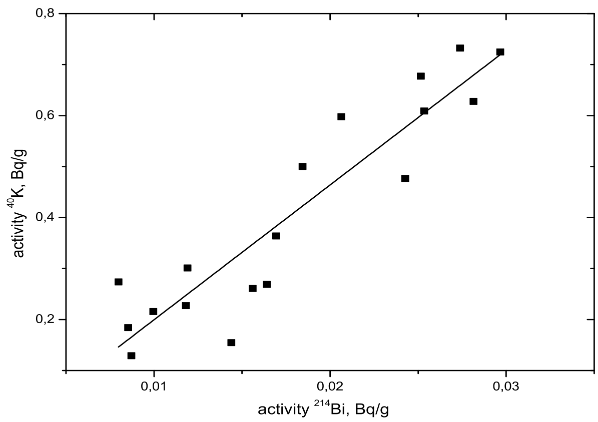

Fig.4. *Dependence of activity of ^{40}K vs ^{214}Bi (^{226}Ra) in the soil of the Kharkiv region*

In Figs.3-5 the dependencies of activity ^{40}K vs ^{238}U, ^{40}K vs ^{214}Bi (corresponds ^{226}Ra) and ^{40}K vs ^{208}Tl (corresponds ^{232}Th) in soil of the Kharkov region are given. Approximation of the above dependencies is carried out by a linear function. It is possible to see that the most smaller straggling of the values concerning a linear function is observed for dependence ^{40}K/^{232}Th. As stated above, it is caused by high fastness of thorium minerals in the soil. Small enough straggling of value is observed and for dependence ^{40}K/^{214}Bi. We will notice that points values of ^{40}K which strongly differ from a linear dependence, correspond to samples which had been taken from an arable land. The greatest straggling of values is realized for dependence ^{40}K/^{238}U. In general, uranium is more closely associated with iron and manganese, due to its strong sorption by iron oxides. Radium, on the other hand, is more related to Ca and Ba in its behavior [4].

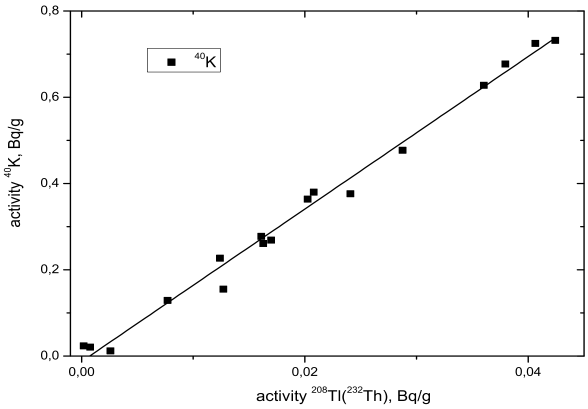

Fig.5. *Dependence of activity of ^{40}K vs ^{208}Tl (^{232}Th) in the soil of the Kharkiv region*

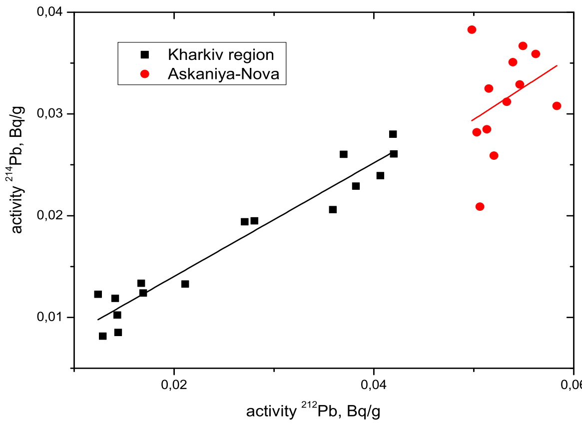

Fig.6. *Dependence of activity of ^{214}Pb vs ^{212}Pb in the soil of the Kharkiv region and Askaniya-Nova*

The intensity of migration of radioactive elements by of water under oxidizing conditions forms the following series: ^{238}U $>$ ^{226}Ra $>$ ^{232}Th. Due to the different migration mobility of natural radioactive elements, a redistribution of these elements between the conjugate landscapes is observed. Thus, U(VI) is removed from eluvial landscapes and concentrates on geochemical barriers such as floodplain, marsh and swampy soils, where it is restored to a relatively inactive U(IV). In humid eluvial and trans-eluvial landscapes, soils are depleted of uranium in favor of radium in comparison with their equilibrium ratio. Hydromorphic soils of accumulative landscapes are enriched uranium. In alluvial sediments, the concentrations of U and Ra are low due to their leaching and removing by water. The ratio Ra/U in alluvial sediments is high since U is more mobility and is predominated in the water. The shift of radioisotope ratios in soils of conjugate landscapes is observed for Th/U and Th/Ra since thorium is characterized by much lower migration mobility in comparison with U and Ra [8]. For soils of the humid zone, the migration series of elements Ra $>$ U $>$ Th were determined.

In our case, Ra is more mobile, which confirms the humid climate of the Kharkiv region [4] (see Fig.2).

The dependence of ^{214}Pb vs ^{212}Pb, which is related to the ratio of uranium vs thorium and in a greater degree to the ratio of radium vs thorium, is shown in Fig.6. In our case, a significant amount of radium is washed out by water (see Fig.2). Therefore, it can be assumed that a significant amount of radium is found in minerals containing thorium. A certain confirmation of this is the dependence of ^{214}Pb vs ^{212}Pb for the Askaniya-Nova soil (see Fig.6), in which an analogous dependence take place only for large values of ^{212,214}Pb.

The dependence of ^{40}K vs ^{214}Pb shows In Fig.7 (^{214}Pb is the decay product of $^{238}U \rightarrow {}^{226}Ra$). More reliably, this dependence is due to ^{226}Ra. The content of ^{238}U and ^{40}K is greater for the dark chestnut soil of Askaniya-Nova than for chernozems [9]. The ^{40}K content is especially great in the soil of Askaniya-Nova (see Fig.7).

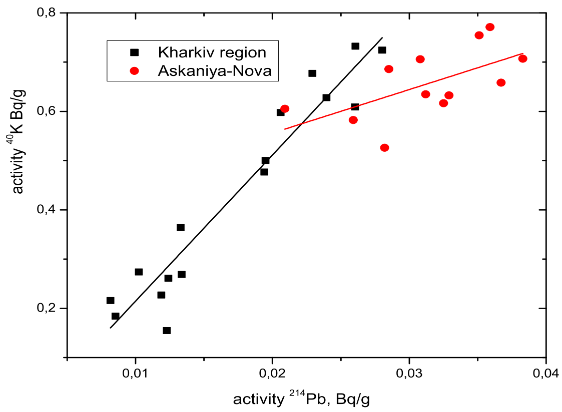

Fig.7. *Dependence of activity of ^{40}K vs ^{214}Pb in the soil of the Kharkiv region and Askaniya-Nova*

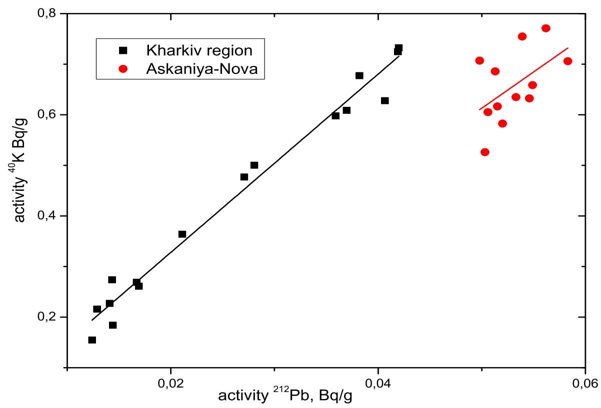

Fig.8. *Dependence of activity of ^{40}K vs ^{212}Pb in the soil of the Kharkiv region and Askaniya-Nova*

The dependence of ^{40}K vs ^{212}Pb is shown in Fig.8 (^{212}Pb product of the decay of ^{232}Th). The mobility of Th in soil is low, so the dependence of ^{40}K vs ^{212}Pb for the soil of Askaniya-Nova is similar it's for the soil of Kharkiv region, which corresponds to the conventional value of thorium content in the dark chestnut soil [10].

The dependence of ^{40}K vs ^{137}Cs is shown in Fig.9. It can be seen that there are no correlation links. It can be assumed that the ^{137}Cs content is due to its sorption by illite of soil. Due to water erosion, the caesium moves from the humid area or it is transported to deeper horizons of the soil.

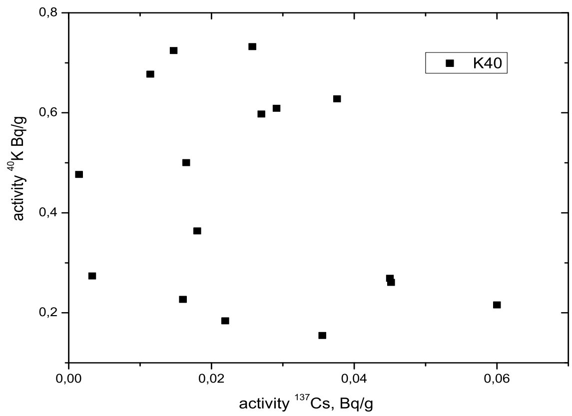

Fig.9. *Dependence of activity of ^{40}K vs ^{137}Cs in the soil of the Kharkiv region*

4. CONCLUSIONS

1. The series of migration elements (Ra > U > Th) in the soil of the Kharkiv region is determined by gamma spectrometer method. These series of migration elements corresponds to the humid type of climate. A high mobility of radium as respect to uranium and thorium has been found.

2. A high correlation between ^{40}K vs ^{232}Th was found. Therefore, as a result of low mobility of thorium, it is possible to assume that thorium and potassium enter into the composition of the mother minerals.

3. Correlation of ^{137}Cs with other radioactive isotopes was not significant in the soil of the Kharkiv region. This apparently indicates specifies in that a high mobility clay minerals can sorbing of cesium.

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СОДЕРЖАНИЕ ПРИРОДНЫХ РАДИОАКТИВНЫХ ИЗОТОПОВ В ПОЧВЕ ХАРЬКОВСКОЙ ОБЛАСТИ

Н.П. Дикий, Ю.В. Ляшко, Е.П. Медведева, Д.В. Медведев, Ю.Г. Пархоменко, И.Д. Федорец

Содержание и миграция природных радиоактивных изотопов в некоторых участках поверхностной почвы Харьковской области изучались с помощью низкофоновой гамма-спектроскопии. Определена миграционная серия радиоактивных элементов (Ra $>$ U $>$ Th) в почве Харьковской области, что соответствует гумидному типу климата. Обнаружена высокая подвижность радия по отношению к урану и торию. Обнаружена высокая корреляция между ^{40}K vs ^{232}Th, которая подтверждает, что торий и калий входят в состав материнских минералов из-за низкой подвижности тория. Корреляция ^{137}Cs с другими радиоактивными изотопами не существенна в почве Харьковской области. Это, по-видимому, указывает на то, что высокомобильные глинистые минералы могут сорбировать цезий.

ВМІСТ ПРИРОДНИХ РАДІОАКТИВНИХ ІЗОТОПІВ У ГРУНТІ ХАРКІВСЬКОЇ ОБЛАСТІ

М.П. Дикий, Ю.В. Ляшко, О.П. Медведева, Д.В. Медведєв, Ю.Г. Пархоменко, І.Д. Федорець

Вміст і міграція природних радіоактивних ізотопів у деяких ділянках поверхневого грунту Харківської області вивчалися за допомогою низькофонової гамма-спектроскопії. Визначена міграційна серія радіоактивних елементів (Ra $>$ U $>$ Th) у грунті Харківської області, що відповідає гумідному типу клімату. Виявлена висока рухливість радію по відношенню до урану і торію. Виявлена висока кореляція між ^{40}K vs ^{232}Th, яка підтверджує, що торій і калій входять до складу материнських мінералів через низьку рухливость торію. Кореляція ^{137}Cs з іншими радіоактивними ізотопами не є суттєвою в грунті Харківської області. Це, мабуть, вказує на те, що високомобільні глинисті мінерали можуть сорбувати цезій.