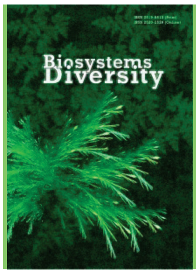

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Geoinformation modeling of radioactive contamination of territories on the example of mines of the "ShidGSK" mining and processing plant

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The aim of the study was to model the territorial distribution and statistical assessment of migration through the trophic chain of naturally occurring radionuclides released into the environment as a result of uranium ore mining. Statistical, geoinformation and experimental research methods were used in the study. Interpolation of the results of volumetric activity of natural radionuclides in environmental components and development of spatial models of their territorial distribution were performed using the ArcGIS software; statistical processing of modelling results and development of mathematical models of migration of natural radionuclides between environmental components were performed using ArcGIS Geostatistical Analyst software. The paper substantiates the choice of the method of geostatistical modelling of the territorial distribution of volumetric activity of natural radionuclides ^{234}U, ^{238}U, ^{210}Po, ^{210}Pb, ^{226}Ra in soils and plants, which allows modelling the values of probabilistic indicators of radioecological contamination in the absence of a sufficient array of initial actual research results. Based on the analysis of the data obtained, the methodology of mathematical modelling of migration of natural radionuclides between soil and plant parts was further developed, which will facilitate consideration of the specifics of migration of natural radionuclides through the trophic chain and help in determining the level of radioecological hazard to the environment. The developed territorial models allow one to obtain stochastic data for their further analytical processing and visualizing radioecological hazard zones. Based on the developed models, zones of increased radioecological hazard within the existing sources of radioactive contamination were identified.

Keywords: natural radionuclides; volumetric activity; soil; vegetation; root crops; interpolation; modeling; correlation.

Introduction

The problem of radioactive contamination of individual regions of Ukraine and the country as a whole has remained relevant in recent decades, not only due to the large-scale accident at the Chornobyl nuclear power plant, but also due to the utilization of nuclear weapons and used nuclear power plant components. Since the mid-twentieth century, uranium deposits have been developed and enriched in some regions of Ukraine, which has resulted in the accumulation of significant amounts of radioactive waste that has a dangerous impact on the ecosystem of the surrounding areas, as well as on the health of the population in these regions.

An example of radioactive materials storage is the tailings storage facility "Dniprovske" (the city of Kamianske, Ukraine), where about $5{,}85 \cdot 10^6$ m^3 of hazardous waste was accumulated in 1954–1968 on an area of about 76 ha in the floodplain of the Dnipro river. The lack of an adequate waterproof screen under the tailing pit and earthen dam leads to constant flooding of radioactive materials, their leaching and migration in groundwater to the nearest small river Konoplyanka (Rudakov et al., 2023).

Another radiation-contaminated region of Ukraine is Kirovohrad Oblast, and in particular the area surrounding the city of Kropyvnytskyi, where the State Enterprise "Eastern Mining and Processing Plant" has been operating since 1951, developing uranium ore deposits and performing the full range of works on the extraction and radiometric enrichment of uranium ores (State Enterprise "Eastern Mining and Processing Plant," 2023).

Thus, at the stages of extraction and enrichment of mineral raw materials, natural radionuclides are released into the environment through emissions and discharges from enterprises (Kutsak et al., 2017b; Soroka, 2021).

Natural radionuclides are released into the environment by emissions and discharges of enterprises already at the stages of extraction and enrichment of mineral raw materials. In particular, Deforge et al. (2021) notes that all waste rocks of the Ingulska mine (the city of Kropyvnytskyi, Ukraine) are characterized by a uranium content exceeding 0.01%. Deforge et al. (2021) notes that the natural radioactive background is exceeded by two times at a range of 250–300 meters from the location of the dumps and even at a range of 1500 meters from the dumps. Similar observations were noted by scientists for both spontaneous and organised storage of radioactive materials (Rudakov et al., 2023; Heracleous et al., 2024). The data presented in (Wan et al., 2024) indicate the spread of radioactive particles over long distances and in concentrations significantly exceeding background values. The results showed that the maximum U content in the analysed soils reached 84.2 mg/kg. These values are significantly higher than background values for soils in China and other countries.

At mining and enrichment enterprises of non-uranium industries, the sources of constant release of aerosols of natural radionuclides into the atmosphere and natural radionuclides into surface and groundwater are overburden and industrial waste dumps, as well as tailing ponds of enrichment enterprises (Soroka, 2021).

Natural radionuclides do not decay in soil and water but migrate through the trophic chain: "soil → plant (feed) → animal → product → human".

The results of experiments (Wasserman et al., 2024) indicate that more than 80% of the total ^{90}Sr content in soils remains potentially mobile, mainly in the bioavailable phase, more than two years after soil contamination. According to Wasserman et al. (2024), the behaviour of this radionuclide in soils is controlled by ionic competition mechanisms for absorption and sorption sites by roots, stable Sr and basic nutrients (Ca, Mg and K), as well as the mineralogical composition of the soil, as specified (Suárez-Navarro et al., 2023). The high rate of migration of ^{90}Sr down the soil profile indicates a high rate of transfer to groundwater. Groundwater, according to Naresh Tanwer and others (Tanwer et al., 2024), is the main source of uranium intake in the human body, accounting for 85%, while food accounts for 15%.

However, the main share of radionuclides is located in the surface and root layer of the soil, and their movement to deeper horizons is very slow. This leads to the intake of radionuclides into crop products (Alves et al., 2022; Wan et al., 2024; Wasserman et al., 2024), and then into animals and human organisms. Thus, Borshchenko (2009) and Borshchenko & Slavov (2016) identify factors that affect the intensity of ^{137}Cs accumulation by forage species of natural lands with their subsequent use by cattle and proves the dependence of milk quality on the feed base of cattle. It has been determined (Samad et al., 2024) that the mean activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K are 3.08, 8.37, and 253 Bq/kg in soil, 0.50, 0.39, and 203.05 Bq/kg in grass, and 0.29, 0.08, and 29.69 Bq/L in milk, respectively. The values of the transfer coefficient for ^{232}Th, ^{226}Ra and ^{40}K from soil to grass were 0.18, 0.05 and 0.84, respectively. For grass to milk, the values of the transfer coefficient for ^{232}Th, ^{226}Ra and ^{40}K were 0.45, 0.17 and 0.11, respectively. A study by Rozputniy et al. (2019) determined that the transfer coefficients of ^{137}Cs and ^{90}Sr to grain vary widely depending on the type of crop and range from 0.01 to 0.40. This leads to the transfer of up to 0.9% of ^{137}Cs and 0.2% of ^{90}Sr from the daily feed into milk. Accordingly, 10% and 2% of these radionuclides are released with the daily milk yield consumed by the population.

As a result, there occur hidden negative alterations in the overall metabolism in the human body (Lopatyuk, 2020). The main target organs affected by the consumption of radioactively contaminated food are kidneys, bones, lungs, etc. It can cause kidney failure, impaired cell function and bone growth, and DNA mutations (Tanwer et al., 2024). Consumption of products contaminated with radionuclides by the humans results in additional internal exposure of the human body above the natural level. Nevertheless, their content in food and drinking water should not exceed the accepted annual effective internal dose of up to 1 mSv/year (Radiation Safety Standards of Ukraine NRBU-97, 1997).

It should be noted that the current legislation establishes permissible concentrations of uranium radionuclides in drinking water (for the population) (On Approval of Permissible Levels of ^{137}Cs and ^{90}Sr Radionuclides in Food and Drinking Water (DR-97), 1997; Radiation Safety Standards of Ukraine; Supplement: Radiation Protection from Sources of Potential Exposure NRBU-97/D-2000, 2000; Radiation Safety Standards of Ukraine NRBU-97, 1997). However, limiting internal exposure of the population conditionally residing on the border of the sanitary protection zone of uranium mining or processing facilities is achieved by assessing individual doses based on analyses of food samples grown in these areas and reference consumption volumes (Pavlenko, 2010, 2018; Kutsak et al., 2017; Kocher, 2021). It should be noted that inhalation of radionuclides should be considered during assessment of individual internal exposure doses of the population living near such facilities (Kutsak, 2016).

Thus, Voytsitskyi et al. (2019) focus on the ways of foliar and root accumulation of ecotoxicants by plants. They propose a method of chamber models to describe the migration pathways of ecotoxicants in the agroecosystem, where the migration of substances between chambers is determined by the transition coefficients. However, a review of studies describing methods (Medved' & Černý, 2019) for modelling radionuclide transport shows a wide range of options. Such methods include modelling based on balance equations, values of key transfer parameters (diffusion coefficients), and experimental methods that can be used to determine them.

Thus, in areas with levels of contamination at which agricultural activities are possible, although the exposure dose does not exceed the established limit, a significant proportion of the population is exposed to low regular doses, which increases the likelihood of long-term radiobiological repercussions (development of tumours, mutations, and reduced immunity) (Lopatyuk, 2020; Deforge et al., 2021; Tanwer et al., 2024). The purpose of the study was to model the territorial distribution and statistical assessment of the migration through the trophic chain of natural radionuclides released into the environment as a result of uranium ore mining.

Materials and methods

Study area and data collection. To achieve this goal, the following research objectives have been set:

– to perform an analysis of the state of pollution of environmental components within the research object;

– to develop models of the territorial distribution of natural radionuclides in the soils and plants of the study area using geographic information modelling tools;

– to conduct mathematical modelling of migration of natural radionuclides between environmental components;

– to justify the need to increase the level of radioecological safety of the population living within the territory of operating uranium mines.

The object of research is the processes of migration and territorial distribution of natural radionuclides in the environment.

The subject of the study is the level of radiation contamination of the environment and methods of modelling the migration of natural radionuclides between environmental components.

Statistical, geoinformation and experimental research methods were used in the paper. Interpolation of the results of volumetric activity of natural radionuclides in environmental components and development of spatial models of their territorial distribution were performed using the ArcGIS 9.2 software (ESRI, USA); statistical analysis of modelling results and development of mathematical models of migration of natural radionuclides between environmental components were performed using the ArcGIS 9.2 Geostatistical Analyst computer programs (ESRI, USA).

The scientific novelty of the results of the research is described below:

– for the first time, the choice of the method of geostatistical modelling of the territorial distribution of the volumetric activity of natural radionuclides in soils and plants was substantiated, which allows modelling the values of probabilistic indicators of radioecological contamination in the absence of a sufficient array of initial actual research results;

– the methodology for mathematical modelling of migration of natural radionuclides between soil and plant parts was further developed, which will make it possible to consider the specifics of migration of natural radionuclides through the trophic chain and determine the level of radioecological hazard to the environment.

Results

Characterization of the research object and radioecological situation within its boundaries. The industrial sites of the Pivdenna and Pivnichna mines of the State Enterprise "Eastern Mining and Processing Plant" (the city of Kropyvnytskyi, Ukraine) and the territories of the villages of Pervozvanivka, Zavadivka, Neopalymivka, Sonyachnyi, Hirskyi (Kirovohrad region, Ukraine), which fall within the range of influence of these mines and waste rock dumps after uranium ore enrichment, were chosen as the object of this study.

Samples of soil, aerial parts of plants (perennial grasses) and roots of agricultural plants (potatoes, beets, carrots) were analysed within the specified area to assess the volumetric activity of natural radionuclides (^{234}U, ^{238}U, ^{210}Po, ^{210}Pb, ^{226}Ra). We have analysed 18 soil and plant samples (10 of which were taken from the sanitary protection zone of the enterprise), as well as 10 samples of root crops grown on the gardens of local residents (Fig. 1).

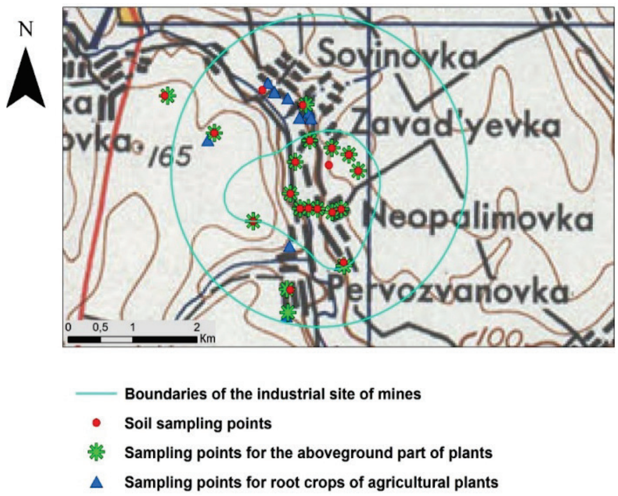

Fig. 1. Map of the research object and sampling sites

Samples. To compare the results, similar samples were analysed at two background locations within a 30-kilometer distance from the mine sites (Velyka Vyska and Subottsi villages, Kirovohrad region, Ukraine).

The measurement results indicate fluctuations in the volumetric activity of natural radionuclides in the soils around the dumps and the mine site, with the highest values in the soils within the ore processing plant of the Pivdenna mine site, which are twice as high as those around the dumps located within the sanitary protection zone of the mines. The radioecological situation within the dumps is characterized by a twofold excess of background values of volumetric activity of all natural radionuclides.

Within the research area, at the place of residence of critical population groups living in the nearest villages, for all observation points, a significant excess of background values was recorded for the following:

– isotopes of uranium, lead and polonium within the villages of Neopalymivka and Pervozvanivka, which are located to the south almost within the sanitary protection zone of the Pivdenna and Pivdenne mines;

– isotopes of radium within the Zavadivka village to the north of the sanitary protection zone of the Pivdenna and Pivdenna mines.

The highest volumetric activity compared to background values is observed in plant samples taken within the dumps for all natural radionuclides studied, except for lead. For all natural radionuclides, background values were exceeded.

According to the results of measurements of volumetric activity of natural radionuclides in root crop samples, it was found that potato samples have the highest volumetric activity compared to beet samples, with a double excess for uranium, polonium and lead isotopes, in particular for Pervozvanivka village. At the same time, the reverse trend is observed for the same radionuclides in Zavadivka village.

Modelling of territorial distribution of natural radionuclides in environmental components using GIS software. As indicated on the map (Fig. 1), the observation points for the environmental components under study do not always overlap spatially, which makes it impossible to determine the correlation between them and predict the spatial distribution of the studied natural radionuclides. The task of forecasting is also complicated by the small amount of input data and a significant difference in the obtained results of the volumetric activity of natural radionuclides (dumps – sanitary protection zone – observation zone – background).

Considering the rather high population density in the observation area, as indicated on the map in Figure 1, and the insufficient territorial and quantitative coverage of sampling, it is planned to model the surfaces of the territorial distribution of natural radionuclides within the study area using the basic set of interpolation tools of ArcGIS software (Natural neighbour interpolation, Inverse distance weighted (IDW), Kriging, Spline).

The selection of modelling methods with different settings was carried out empirically, however, the Natural neighbour interpolation method was found to be the most optimal way to interpolate these data under the conditions of the initial parameters. The surface obtained in this way makes it possible to smooth out the peak values of the indicators and defines a wide gradient that is more similar to the natural distribution, while the background values of the indicators also have a significant impact on the surface modelling.

Discussion

Figure 2 shows the result of surface modelling interpolating the values of ^{234}U volumetric activity in soil, plants, and roots. The constructed surfaces allowed us to model the values of volumetric activity of natural radionuclides in soils and parts of plants for any point of the study area and to visualize the territorial distribution of radioactive contamination.

The constructed surfaces made it possible to model the values of the volumetric activity of natural radionuclides in soils and plant parts for any point of the study area and to visualize the territorial distribution of radioactive contamination.

According to the modelling maps interpolating the volumetric activity of natural radionuclides in the soil, the epicentre of contamination is observed within the waste rock dumps located within the southwestern border of the sanitary protection zone of the mine site and is more than 10 times higher than the background value for uranium and radium isotopes, up to 20 times higher for lead and polonium isotopes within the village of Pervozvanivka.

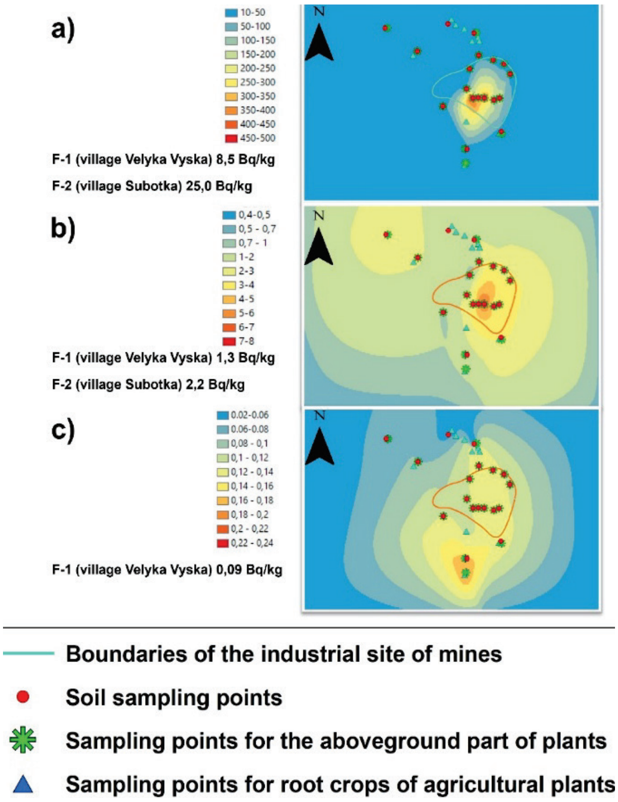

Fig. 2. Map of surface modelling by the method of Natural neighbour interpolation, which interpolates the results of bulk activity on the example of ^{234}U (Bq/kg): *a* – in soil; *b* – in plants; *c* – in root plants

Modelled indicators of volumetric activity of natural radionuclides in the aboveground part of plants indicate a tenfold excess of background values within the sanitary protection zone of the mine site; a twofold excess of background values of uranium and lead isotopes was found within the villages of Hirskyi, Neopalymivka and Pervozvanivka, respectively.

It has been determined that the epicentre of radioactive contamination of agricultural plant roots is observed within the settlements of Neopalymivka and Pervozvanivka with an excess of background values by 2.0–2.5 times, and the zones of lead isotope contamination with a twofold excess of background values extend to the settlements of Sonyachnyi and Hirskyi.

The developed digital maps of radionuclide distribution will allow a comprehensive approach to assessing the risk of hazard to the population living within the source of radioactive contamination and modelling the distribution of radioactive contamination in agricultural plants, taking into account their consumption by the local population, will allow developing recommendations for agri-cultural activities within this area.

Mathematical modelling of migration of natural radionuclides between environmental components. The constructed surfaces shown in Figure 2, made it possible to model the volumetric activity of natural radionuclides for soils, plants, and root crops at any point of the study area. By creating a uniform distribution of virtual study points, the result of which is shown in Figure 3, the values of the volumetric activity of natural radionuclides for soils, plants, and root crops were obtained at each of the 300 specified points.

It is worth noting that the territorial coordinates of the modelled points for all environmental components coincide, and therefore are available for determining the pairwise correlation.

Based on the methods of mathematical statistics using Geostatistical Analyst computer software (ESRI, USA), correlations between the values of volumetric activity of natural radionuclides modelled for virtual points in soil and plant elements were established. The results of mathematical modelling for the example of ^{234}U are shown in Figure 4, and using the Geostatistical Analyst toolkit – in Figure 5.

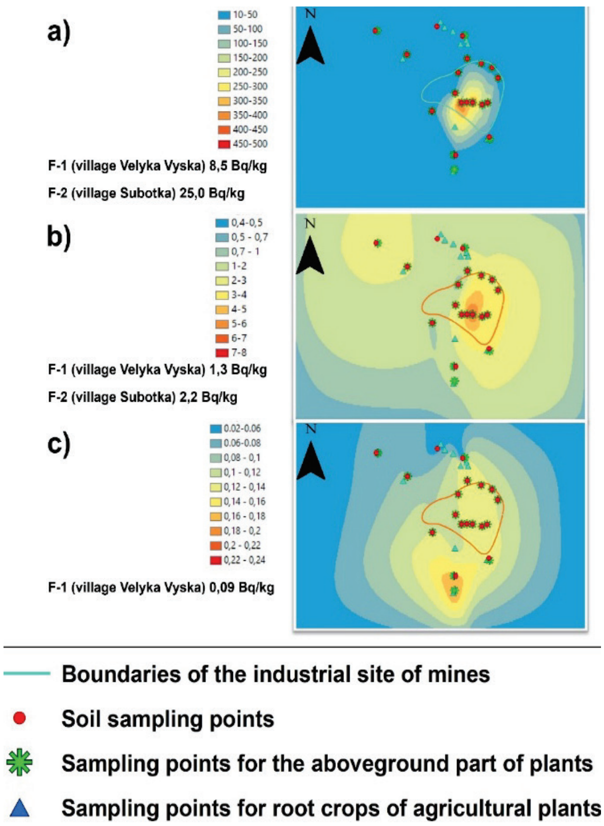

Fig. 3. Map of creating a uniform distribution of virtual research points

The models show that the closest relationship is observed between the values of ^{234}U volumetric activity in soils and the aerial parts of plants, as indicated by the high correlation coefficient (R = 0.87) and high density of mutual covariance of the soil/plant pair model.

The models were built in a similar way for all natural radionuclides studied. The modelling results are shown in Table 1. The obtained pairwise correlation models indicate a high correlation (R = 0.78–0.87), primarily of a logarithmic nature, of such parameters as the value of the volumetric activity of natural radionuclides in soils and aboveground parts of plants, in particular for uranium isotopes, as well as ^{210}Po and ^{226}Ra. For ^{210}Pb, this dependence was derived with a low coefficient of determination, indicating the presence of other factors that are more important.

Regarding the dependence of the volume activity of natural radionuclides in root crops on the indicators modeled for soil samples, a much lower correlation than in the previous case (R = 0.48–0.71) was found to be mainly polynomial.

Justification of the obtained results. The obtained modelling results confirm the reliability of the chosen method for modelling the process of migration of natural radionuclides through the trophic chain, but the determined parameters of mathematical models indicate a more likely accumulation in the surface soil layer accessible to the root system of perennial grasses, which have primarily a superficial root system and serve as a kind of protection of deep soil horizons from the movement of pollutants, accumulating a significant amount of natural radionuclides in their green mass.

According to the results obtained for the soil/root crops parameter pair, the movement of natural radionuclides to deeper soil horizons is significantly slower under these natural conditions, which leads to less active radionuclide intake into crop products (root crops) than into the aboveground green mass of plants.

The results of modelling can be used in the field of radioactive waste management to predict long-term behaviour and assess safety standards. The developed mathematical models of migration of natural radionuclides between soil and plants, given their high reliability, can be used to predict the levels of radioecological contamination of agricultural products grown by local residents in their household plots. And also for the development and application of rehabilitation measures similar to those that were developed for the territories affected by the consequences of the accident at the Chornobyl NPP (Ukraine), at the Fukushima Daiichi nuclear power plant (Japan). For example, groups of authors (Onda et al., 2020; Fesenko et al., 2021) summarized information on the application of control measures after the accident, analyzed the experience of applying agricultural rehabilitation measures and their impact on the radiological situation in different periods after the accident, and evaluated their effectiveness.

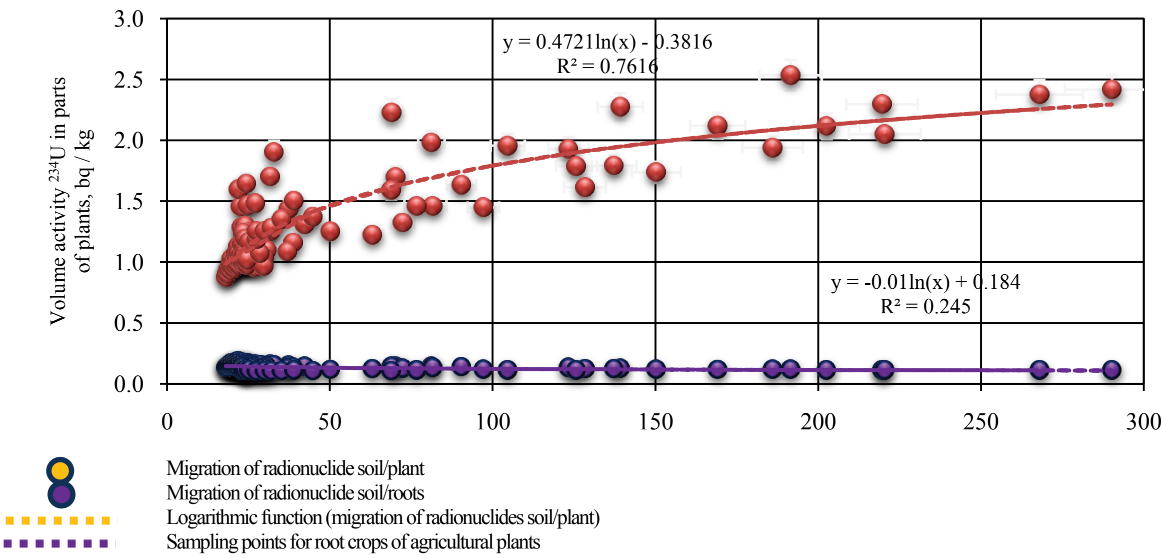

Fig. 4. Mathematical model that characterizes the probability of migration of natural radionuclides between environmental components on the example of ^{234}U (Bq/kg)

Further calculations of the volumes of consumption of agricultural products contaminated with radionuclides grown in the contaminated area will allow the risks of additional internal exposure of the human body to be determined. And taking into account possible trophic chains:

"Soil → roots → human"

"Soil → perennial grasses → animal → livestock products → human"

will allow recommendations to be provided on the norms of consumption of these agricultural products or the possibility of conducting agricultural activities within the territory in general, since regular exposure, albeit at low doses, increases the likelihood of radio-biological effects in the local population. As noted Kocher (2021), the introduction of standards for the protection of the population from the harmful effects of ionising radiation will limit the risk (probability) of stochastic health effects, including cancers, leukaemias and severe hereditary effects, and will prevent non-stochastic or deterministic effects in organs or tissues.

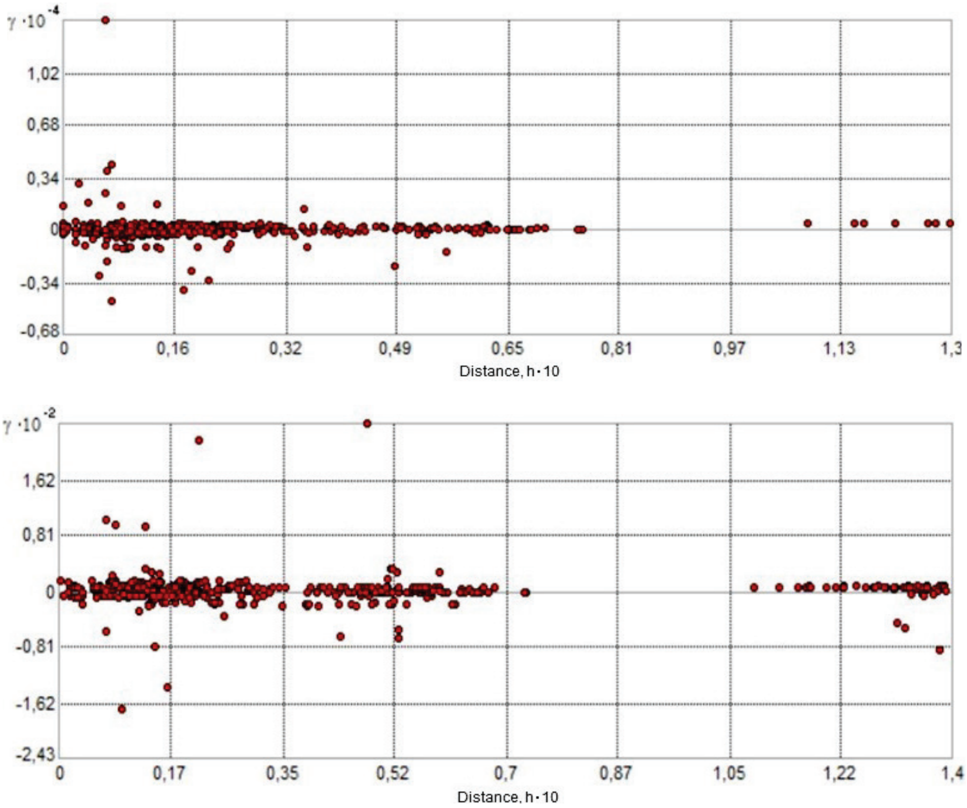

Fig. 5. Statistical covariance model characterizing the probability of migration of natural radionuclides between environmental com-ponents on the example of ^{234}U (Bq/kg): *a* – pair model soil/plants; *b* – pair model soil/root crops

Table 1
Results of mathematical modelling of migration of natural radionuclides between soil and plants

Natural radionuclides	Soil / Plant	Soil / Roots
^{234}U	$y_1 = 0.4721\ln(x) - 0.3816$ $R^2 = 0.7616$	$y_2 = -0.013\ln(x) + 0.1846$ $R^2 = 0.245$
^{238}U	$y_1 = 0.4146\ln(x) - 0.3178$ $R^2 = 0.7298$	$y_2 = 0.0005x^2 - 0.0286x + 0.4324$ $R^2 = 0.3666$
^{210}Pb	$y_1 = -0.331x^2 + 19.01x - 261.22$; $R^2 = 0.2658$	$y_2 = -0.0018x^2 + 0.1082x - 1.4305$; $R^2 = 0.295$
^{210}Po	$y_1 = 68.985e^{-0.144x}$ $R^2 = 0.6352$	$y_2 = 0.0067x^2 - 0.3033x + 3.4542$ $R^2 = 0.23$
^{226}Ra	$y_1 = 0.458\ln(x) - 0.2344$ $R^2 = 0.6055$	$y_2 = -0.0006x^2 + 0.0292x - 0.271$ $R^2 = 0.5019$

Note: x is the value of the volume activity of natural radionuclides in soils: y_1 is the value of the volume activity of natural radionuclides in vegetation; y_2 is the value of the volume activity of natural radionuclides in root crops.

Conclusion

Analysis of the state of pollution of environmental components within the industrial site of the Pivdenna and Pivdenne mines of the State Enterprise "Eastern Mining and Processing Plant" and the territory of the surrounding villages (Kirovohrad region, Ukraine) was carried out and a significant excess of volumetric activity of natural radionuclides relative to background values was recorded in soil samples and plant elements for uranium isotopes, lead and polonium isotopes within the settlements located south of the sanitary protection zone of the mine site, and radium isotopes – to the north of them.

The models of the territorial distribution of natural radionuclides in the soils and plants of the study object were developed using the basic set of interpolation tools of ArcGIS software. It has been established that the optimal method of geospatial modelling is the Natural neighbor interpolation method, which makes it possible to smooth out the peak values of indicators and determines a wide gradient, taking into account the significant influence of background values of indicators. The constructed surfaces made it possible to model the values of the volumetric activity of natural radionuclides in soils and plant parts for any point of the study area. The visualized territorial distribution of indicators indicates the epicenter of radioactive contamination within the waste rock dumps located within the boundary of the sanitary protection zone of the mine site, 10–20 times higher than the background values of the studied natural radionuclides in soils and 2–10 times higher in plant parts within the adjacent settlements.

The migration of natural radionuclides between environmental components was modeled using mathematical statistics methods with the use of Geostatistical Analyst computer programs, and a close correlation was established between the values of the volumetric activity of natural radionuclides in the soil and aboveground parts of plants, which confirms the reliability of the chosen method for modelling the migration of natural radionuclides through the trophic chain. The modelling results indicate a more probable accumulation of natural radionuclides in the surface soil layer, which slows down with depth to soil horizons accessible to root crops.

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