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 Chernobyl 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 "Dniprovsky" (the city of Kamianske, Ukraine), where about 5.85 x 10^9 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 (Radakov 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 (Radakov 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 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 (Staier-Navarro et al., 2023). The high rate of migration of 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%.

Keywords: natural radionuclides; volumetric activity; soil; vegetation; root crops; interpolation; modeling; correlation.
respectively. The values of the transfer coefficient for 232Th, 226Ra and 40K are 3.08, 8.37, and 0.39, respectively. For grass to soil, the values of the transfer factor coefficient for 232Th, 226Ra and 40K were 0.18, 0.05 and 0.84, respectively. For grass to milk, the values of the transfer coefficient for 232Th, 226Ra and 40K were 0.17 and 0.11, respectively. A study by Rozputnyi et al. (2019) determined that the transfer coefficients of 137Cs and 90Sr 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 137Cs and 0.2% of 90Sr 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 the occurrence of diseases related to bone damage, kidney failure, and cancer (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 the occurrence of diseases related to bone damage, kidney failure, and cancer (Lopatyuk, 2020). 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The main target organs affected by the consumption of radionuclides in drinking water (for the population) (On Approval of Permissible Levels of 137Cs and 90Sr 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; Kucher, 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 ecotoxins by plants. They propose a method of chamber models to describe the migration pathways of ecotoxins 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. 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 radiocological 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 radiocological hazard to the environment.

Results

Characterization of the research object and radiocological 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, Neopalyntovka, Sonyachnyi, Hilskyi (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 (238U, 235U, 232Th, 226Ra, 210Po, 210Pb, 239,240Pu) in the 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).

![Map of the research object and sampling sites](image)

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

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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 radiocological 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 Pivdenne 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 $^{238}$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.

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 modelling results are shown in Table 1. The obtained pairwised 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 determinati- 
on, 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 determin- 
ed parameters of mathematical models indicate a more likely accumula- 
tion 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, accum- 
ulating 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 sig- 
ificantly slower under these natural conditions, which leads to less active 
radionuclide intake into crop products (root crops) than into the abovegro-
und 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 radiocological 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 deve- 
loped 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 af- 
ter the accident, analyzed the experience of applying agricultural rehabili-
tation measures and their impact on the radiological situation in different 
periods after the accident, and evaluated their effectiveness.

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 radiological 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 can- 
cers, 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 components on the example of 234U (Bq/kg): a – pair model soil/plants; b – pair model soil/root crops.

### Table 1

<table>
<thead>
<tr>
<th>Natural radionuclides</th>
<th>Soil/Plant</th>
<th>Soil/Roots</th>
</tr>
</thead>
<tbody>
<tr>
<td>234U</td>
<td>$y_1 = 0.4721\ln(x) - 0.3816$</td>
<td>$y_2 = -0.0013\ln(x) + 0.1846$</td>
</tr>
<tr>
<td></td>
<td>$R^2 = 0.7616$</td>
<td>$R^2 = 0.9245$</td>
</tr>
<tr>
<td>238U</td>
<td>$y_1 = 0.4146\ln(x) - 0.3178$</td>
<td>$y_2 = 0.0005x^2 - 0.0286x + 0.4324$</td>
</tr>
<tr>
<td></td>
<td>$R^2 = 0.7298$</td>
<td>$R^2 = 0.3666$</td>
</tr>
<tr>
<td>210Pb</td>
<td>$y_1 = -0.331x^2 + 19.01x - 261.22$</td>
<td>$y_2 = -0.0018x^2 + 0.1082x - 1.4305$</td>
</tr>
<tr>
<td></td>
<td>$R^2 = 0.2658$</td>
<td>$R^2 = 0.295$</td>
</tr>
<tr>
<td>210Po</td>
<td>$y_1 = 68.985e^{-0.144x}$</td>
<td>$y_2 = 0.0067x^2 - 0.3033x + 3.4542$</td>
</tr>
<tr>
<td></td>
<td>$R^2 = 0.6352$</td>
<td>$R^2 = 0.23$</td>
</tr>
<tr>
<td>226Ra</td>
<td>$y_1 = 0.458\ln(x) - 0.2344$</td>
<td>$y_2 = -0.0006x^2 + 0.0292x - 0.271$</td>
</tr>
<tr>
<td></td>
<td>$R^2 = 0.6055$</td>
<td>$R^2 = 0.5019$</td>
</tr>
</tbody>
</table>

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 (Karvovhrad 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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